PB82-112384

Radon Transport Through and Exhalation from Building Materials: A Review and Assessment

(U.S.) National Bureau of Standards
Washington, DC

Prepared for

Office of Radiation Programs
Washington, DC

Sep 81

U.S. Department of Commerce
National Technical Information Service
NTIS

NBS-114A (REV. 2-80) PB82 112384

U.S. DEPT. OF COMM.
BIBLIOGRAPHIC DATA SHEET (See instructions)

1. PUBLICATION OR REPORT NO.
NBS TN 1139

2. Performing Organ. Report No.

3. Publication Date
September 1981

4. TITLE AND SUBTITLE
Radon Transport Through and Exhalation From Building Materials: A Review and Assessment

5. AUTHOR(S)
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U28103 8

6. PERFORMING ORGANIZATION (If joint or other than NBS, see instructions)
NATIONAL BUREAU OF STANDARDS
DEPARTMENT OF COMMERCE
WASHINGTON, D.C. 20234

7. Contract/Grant No.
AD-13-F-0-133-0

8. Type of Report & Period Covered
Interim

9. SPONSORING ORGANIZATION NAME AND COMPLETE ADDRESS (Street, City, State, ZIP)
Office of Radiation Programs
U.S. Environmental Protection Agency
Washington, DC 20460

10. SUPPLEMENTARY NOTES

☐ Document describes a computer program; SF-185, FIPS Software Summary, is attached.

11. ABSTRACT (A 200-word or less factual summary of most significant information. If document includes a significant bibliography or literature survey, mention it here)

This report was prepared, at the request of the U.S. Environmental Protection Agency, for the purpose of reviewing, assessing, and summarizing what is currently known about radon transport through and exhalation from building materials. In four chapters, the report 1) considers the routes of entry of radon into buildings, describes the basic models for radon transport through building materials, critically reviews the small number of existing values for the necessary transport coefficients, and summarizes the solutions of both steady-state and time-dependent transport cases; 2) reviews and considers how the microstructural properties and internal characteristics of building materials may affect the transport and exhalation of radon; 3) considers the exhalation process from a more macroscopic, phenomenological viewpoint, and summarizes selected experimental data on radium concentrations in building materials, radon flux and exhalation from soils and building materials, and the effects of meteorological variables on radon exhalation; and 4) reviews and assesses various measurement methodologies that are used for laboratory and in situ studies of radon transport and exhalation. Needs for further research in each area are also recommended.

12. KEY WORDS (Six to twelve entries; alphabetical order; capitalize only proper names; and separate key words by semicolons)
buildings; concrete; diffusion; exhalation; materials; measurement; permeability; radon; transport

13. AVAILABILITY
☒ Unlimited
☐ For Official Distribution. Do Not Release to NTIS
☒ Order From Superintendent of Documents, U.S. Government Printing Office, Washington, D.C. 20402.
☐ Order From National Technical Information Service (NTIS), Springfield, VA. 22161

14. NO. OF PRINTED PAGES
101

15. Price
$4.50

☆U.S. GOVERNMENT PRINTING OFFICE: 1981-340-997/1799

USCOMM-DC 6043-P80

PB82-112384

NBS TECHNICAL NOTE 1139

U.S. DEPARTMENT OF COMMERCE / National Bureau of Standards

RADON TRANSPORT THROUGH AND EXHALATION FROM BUILDING MATERIALS:

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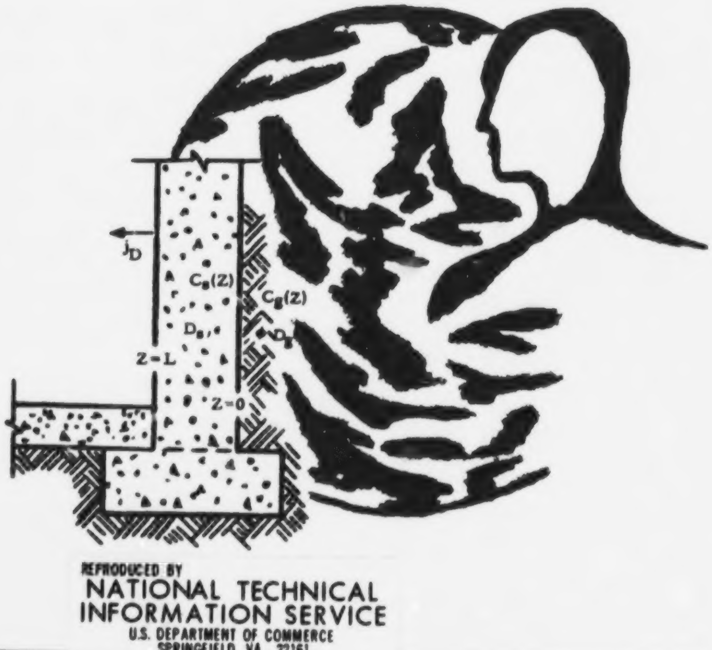

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U.S. DEPARTMENT OF COMMERCE, Malcolm Baldrige, Secretary
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Issued September 1981

National Bureau of Standards Technical Note 1139
Nat. Bur. Stand. (U.S.), Tech. Note 1139, 101 pages (Sept. 1981)
CODEN: NBTNAE

ABSTRACT

This report was prepared, at the request of the U.S. Environmental Protection Agency, for the purpose of reviewing, assessing, and summarizing what is currently known about radon transport through and exhalation from building materials. In four chapters, the report 1) considers the routes of entry of radon into buildings, describes the basic models for radon transport through building materials, critically reviews the small number of existing values for the necessary transport coefficients, and summarizes the solutions of both steady-state and time-dependent transport cases; 2) reviews and considers how the microstructural properties and internal characteristics of building materials may affect the transport and exhalation of radon; 3) considers the exhalation process from a more macroscopic, phenomenological viewpoint, and summarizes selected experimental data on radium concentrations in building materials, radon flux and exhalation rates from soils and building materials, and the effects of meteorological variables on radon exhalation; and 4) reviews and assesses various measurement methodologies that are used for laboratory and in situ studies of radon transport and exhalation. Needs for further research in each area are also identified.

Key words: Buildings; concrete; diffusion; exhalation; materials; measurement; permeability; radon; transport.

ACKNOWLEDGMENTS

This review and assessment was prepared under the direction of a study group consisting of the authors as well as P. W. Brown (NBS Center for Building Technology), W. A. Cassatt and E. H. Eisenhower (NBS Center for Radiation Research), and I. C. Sanchez and L. E. Smith (NBS Center for Materials Science). We want to thank these members of the study group for their contributions to this report. Special thanks are due Robert Schofield, our summer student assistant, who painstakingly and efficiently accumulated and organized our extensive reference collection. His contributions were appreciated long after his brief stay at NBS.

Many other persons contributed by carefully reviewing and commenting on an earlier version. Dr. R. A. Helmuth (Chemical and Physical Research Department, Portland Cement Association) was particularly helpful in reviewing Chapter II and in giving us advice. We also benefited from comments and suggestions by Drs. R. Bruno (U.S. Environmental Protection Agency), Niels Jonassen (Technical University of Denmark), W. W. Nazaroff (Lawrence Berkeley Laboratory), J. R. Noyce (NBS Center for Radiation Research), and A. G. Scott (American Atcon, Inc.); and especially from the valuable suggestions and criticisms by Allan Tanner (U.S. Geological Survey). Many of their comments have been incorporated, but, of course, they share no responsibility for any errors in fact or interpretation that may remain.

Lastly, we gratefully acknowledge the NBS National Measurement Laboratory Text-Editing staff and the NBS Technical Information and Publication Division for their excellent assistance in preparing this report.

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INTRODUCTION

The potential health hazards of radiation exposure to the general public due to radon in buildings has, in the past few years, received increasing attention. This concern about indoor radon[*] has been documented in extenso (UN 77; HEW 79: CoM 80; RPC 80), and substantial efforts are being made to study and understand various aspects of the problem. Although several thousand publications relating to indoor radon exist in the open literature, the prediction of indoor radon concentrations is still in a very rudimentary stage. The reasons for this situation arise from the complex nature of the sources and behavior of radon including its release and subsequent movement from the earth and from other repositories of the radium parent, and the dynamics within indoor atmospheres. Another important factor lies in the fragmentary nature of many previous investigations often conducted under uncontrolled conditions and with measurement methodologies of undemonstrated reliability.

One of the most poorly understood and critically needed areas of research concerns the sources of indoor radon and the relation to radon transport through and exhalation from building materials. An understanding of the transport - exhalation mechanism is necessary in order to predict the sources of radon and routes of entry into buildings. Essentially, what is needed is a phenomenological basis or model that can be used (i) to characterize building materials as sources of radon, and (ii) to characterize the mechanisms of radon transport through building materials. This type of predictive capability does not presently exist for several basic reasons:

1. no description of radon transport has been verified mainly because sufficiently accurate transport coefficients and other data (i.e., diffusion coefficients, permeability coefficients, porosity of the building material, and radon production rate per unit pore volume) are not available; and

2. the role of the detailed intrinsic and microstructural properties of building materials (e.g., porosity, pore size and distribution, water content and state) in influencing the entrainment and transport of radon is not well understood;

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[*] Unless otherwise stated, the term "radon" will be used in this report to refer to the ^{222}Rn isotope, and "thoron" to ^{220}Rn. It should also be mentioned that radioactivity is specified throughout this report in units of a submultiple of the "curie" (e.g., pCi) and not in the SI unit "becquerel" (Bq). This was done for convenience since most, if not all, of the existing radon literature uses the older, special units.

3. the influences of external variables such as meteorological conditions on exhalation rates, while qualitatively understood in most cases, are not sufficiently well quantified.

4. radon measurement methodologies, especially those used in situ ("in the field") are frequently not of adequately proven accuracy.

In short, many of the basic processes are not well understood and existing data are not sufficient to quantitatively predict or estimate the flux or exhalation rate of radon from building materials.

The Environmental Protection Agency requested that the National Bureau of Standards address and investigate the above concerns. This report has been prepared, as a part of this overall effort, for the purpose of reviewing, assessing, and summarizing what is currently known about radon transport through and exhalation from building materials. The approach has been to evaluate the literature to the extent possible within very severe time constraints, and to consult with experts in the field. One must recognize that this review is not exhaustive, but, of necessity, selectively considers only those areas that were believed to be most directly applicable to understanding radon transport and exhalation. It must be emphasized that no NBS-based research was undertaken at this stage to verify or validate existing data and information.

This report has been structured into four chapters corresponding to the main areas of concern. Within each chapter, an attempt has been made to identify those areas requiring further study.

The assessment of transport mechanisms is covered in Chapter I. It considers the routes of entry of radon into buildings, describes the basic models for the transport of radon through building materials, critically reviews the small number of existing values for the necessary transport coefficients, and summarizes the solutions of both the steady-state and time-dependent transport equations for various transport cases.

Chapter II reviews and considers how the microstructural properties and internal characteristics of building materials may affect the entrainment and transport of radon through the pore system of the materials. These characteristics include radium distribution, internal geometry, pore size and distribution, and water content and state. Because concrete and other cementitious materials are the basic components of most buildings, hardened concrete is used as an important and representative example to more fully explore the role of these internal characteristics.

Chapter III reviews and summarizes data on radium concentrations in building materials, radon flux and exhalation rates from soils and building materials, and the effects of meteorological variables on radon exhalation. In effect, it focuses on the radon exhalation process from a more macroscopic, phenomenological viewpoint, and addresses whether it is possible to predict or estimate the radon flux or exhalation rate from building materials based on existing data.

Most of what is known about radon transport through or exhalation from building materials is based upon observations and measurements in laboratory or in situ (field) settings. Therefore, an understanding of these previously employed measurement methodologies is necessary in order to evaluate the applicability and validity of existing transport and exhalation data, as well as to plan future laboratory transport and exhalation experiments. In addition, it is reasonable to presume that any predictive phenomenological model will be based in part on field-based measurements. A review and assessment of various measurement methodologies is provided in Chapter IV.

A number of very specific and specialized terms are used throughout this report. Inasmuch as readers may be unfamiliar with some of these terms, the following glossary is provided.

GLOSSARY

DECAY CONSTANT, λ, is a proportionality constant which relates the number of radioactive events per unit time (dn/dt) to the number of radioactive atoms present (n), i.e.,

$$\frac{1}{n}\left(\frac{dn}{dt}\right) = -\lambda$$

The halflife, which is the length of time (T) required for a given number of radioactive atoms to decrease by half, is related to the decay constant by $T = (\ln 2)/\lambda$.

DIFFUSION (or DIFFUSIVE TRANSPORT), which is governed by Fick's Law, refers to the movement of a gaseous component such as radon through the air-filled pore system of a porous material in which the driving force for the movement is a concentration gradient. For a thin slab of material separating regions in which the concentration of radon is different, Fick's Law states that the diffusive flux density of radon through the slab is proportional to the concentration difference and inversely proportional to the slab thickness.

DIFFUSION COEFFICIENT, D_e, is the effective transport coefficient for diffusive transport, and is the proportionality constant, in Fick's law, that relates the diffusive flux density through a porous material to the concentration gradient.

DIFFUSION LENGTH is a characteristic length, ℓ, associated with diffusion of a radioactive species.

EMANATION POWER, π, for a material is equal to the quotient of the number of radon atoms liberated (n_1) from the material by the number of radon atoms formed (n_0) by radioactive decay of radium in the material in unit time (i.e., fractional escape). In effect, it is just the fraction of radon that is "missing" from the material, and may be very dependent on the shape and state of the material. Note that it is also just equal to the ratio of the radon radioactivity released (A_1) to the radon radioactivity produced (A_0) in the material under steady-state conditions.

$$\pi = \frac{n_1}{n_0} = \frac{A_1}{A_0}$$

It is historically the most unequivocal term, (see, for example, Hahn [Hah 36]), and is probably the most intrinsically fundamental exhalation quantity for a unit element of material. Tanner [Tan 78] notes that the Soviet literature uses the term "coefficient of emanation", and that other equivalent terms include "escape ratio", "escape-to-production ratio", and "percent emanation".

EMANATION RATE, following the definition of emanation power, is just the time rate of change of the **emanation power**. It is given by the quotient of $d\pi$ by dt where $d\pi$ is the change in the emanation power in the time interval dt. It may also be expressed in terms of radioactivity

$$\frac{d\pi}{dt} = \frac{d(n_1/n_o)}{dt} = \frac{d(A_1/A_o)}{dt} .$$

In the absence of external influences, the emanation rate (for a given quantity and type of material) is time invariant. Since the formation of radon is relatively constant compared to the effect of external influences on the rate of release of radon, the emanation rate may be approximated by

$$\frac{d\pi}{dt} \simeq \frac{dn_1/dt}{n_o} = \frac{dA_1/dt}{A_o}$$

Obviously, the concept of an emanation rate is a meaningless concept for an unbounded amount of material (e.g., an open field or concrete wall).

EXHALATION refers to the liberation or release of radon from a material. The use of the term **"emanation"** for this concept is deprecated.

EXHALATION RATE, R, is just the time rate of release of radon from a material, and is normally expressed either in units of the number of radon atoms per unit time (e.g., atoms$\cdot$s^{-1}) or in units of a submultiple of the curie by a suitable unit of time (e.g., pCi$\cdot$s^{-1}). It is either given by dn_1/dt or dA_1/dt where dn_1 is the number of radon atoms released from the material in time dt, and dA_1 is the corresponding radon radioactivity (with decay constant λ).

$$\frac{dn_1}{dt} = \frac{1}{\lambda}\left(\frac{dA_1}{dt}\right)$$

The exhalation rate is not normalized to area, mass, radon content of the material, etc. Obviously, the concept of exhalation rate is applicable to both bounded or unbounded quantities of a material.

FLUID FLOW (or **PERMEATION**), which is governed by Darcy's law, refers to the movement of a fluid through the pore system of a porous material in which the driving force is a pressure gradient. For a thin slab of porous material separating regions maintained at different fluid pressures, Darcy's Law states that the fluid flux density through the porous slab is proportional to the pressure difference and inversely proportional to the slab thickness.

FLUX DENSITY is the net number of atoms which pass through a unit cross-sectional area per unit time. For radioactive species, the number of atoms may be specified in terms of its radioactivity.

PERMEABILITY COEFFICIENT, k, (often termed just **PERMEABILITY**) is the transport coefficient for permeability, and is the proportionality constant, in Darcy's law, that relates fluid flux through a porous material to the pressure gradient.

POROSITY, ε, is a characteristic property of a porous material, which is given by the ratio of the volume of the pore spaces in the material to the total volume of the material.

POROUS MEDIUM is a term used to describe a heterogeneous material in which any given point either lies in a void space or in a solid phase, and in which it is possible to move between widely separated points in the void space without crossing a solid boundary.

RADIUM, as used in this report, refers to the ^{226}Ra isotope which is the radioactive progenitor of radon (^{222}Rn).

RADON, as used in this report, refers to the ^{222}Rn isotope (unless explicitly stated otherwise); **thoron** is used to refer to the ^{220}Rn isotope.

RADON FLUX DENSITY, E, (or **AREAL EXHALATION RATE**) for the surface of a material (bounded or unbounded) is the quotient of dR by da where dR is the exhalation rate from the surface of the material with area da.

$$E = \frac{dR}{da}$$

It is just the exhalation rate normalized to unit area, and is normally expressed either as the number of radon atoms per unit area and time (e.g., $\text{atoms}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$) or in a submultiple of the curie by a suitable unit of time and a suitable unit of area (e.g., $\text{pCi}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$).

TRANSPORT is used generically to refer to any mechanism by which radon moves through or from a material.

UNDERPRESSURE (OVERPRESSURE) refers to a pressure difference measured with respect to atmospheric pressure.

VENTILATION RATE (or **AIR EXCHANGE RATE**) is a term applied to a room or a building and is the number of times the air in a unit volume is replaced per unit time.

CHAPTER I

ASSESSMENT OF TRANSPORT MECHANISMS

The purpose of this Chapter is to assess critically the current state of knowledge of radon transport through and exhalation from building materials.

Before proceeding, however, it may be useful to consider the role of transport of radon in or through building materials in the broader context of its contribution to the indoor radon concentration. The radon flux density from the indoor surfaces of a building constitutes only one of several sources of indoor radon. For a closed space such as a room in a building, a theoretical correlation may be established between radon concentration and radon input from various sources. The basis for this correlation is the following balance equation for the different source contributions to the rate of change of the indoor radon concentration [UN 80],*

$$\frac{dC_i(t)}{dt} = (ES/V) + (q/V) + (\alpha_o C_o' + \alpha_s C_s')\lambda_v - C_i(t)(\lambda + \lambda_v) \ , \qquad (1.1)$$

where $C_i(t)$ is the radon concentration in a room at time t;

E is the radon flux density from unit surface of the room;

S is the exhaling surface area of the room;

V is the volume of the room;

q is the radon release rate from a source in the room (water, natural gas);

C_o' is the radon concentration in the inlet or ventilating air;

C_s' is the radon concentration in soil gas which might be drawn into a basement room through, for example, a major crack or a hole in the basement envelope which serves as an entry port for utilities;

α_o and $\alpha_s = 1 - \alpha_o$ are the relative weights of these two external air components of the building ventilation;

λ_v is the ventilation rate (air changes per unit time); and

λ is the radon decay constant.

Under steady conditions in which the source terms remain constant in balance eq. (1.1),

$$\frac{dC_i}{dt} = 0 \ ,$$

---

*This equation, adapted from one appearing in a draft of the revised UNSCEAR report [UN 80], has been modified slightly to suit our present purpose. It should be emphasized that the relative weights, α_o and α_s, which we have introduced may be a function of the ventilation rate.

and the steady-state concentration of radon is

$$C_i = \frac{(ES/V) + (q/V) + (\alpha_o C'_o + \alpha_s C'_s)\lambda_v}{\lambda_v + \lambda} \qquad (1.2)$$

In a typical house, λ_v, which lies in the range $0.1 < \lambda_v < 3\ h^{-1}$, is much greater than the decay constant of radon ($\lambda = 0.01\ h^{-1}$), so

$$C_i \cong (ES/V\lambda_v) + (q/V\lambda_v) + \alpha_o C'_o + \alpha_s C'_s \ . \qquad (1.3)$$

The draft UNSCEAR report [UN 80] lists the relative significance of radon sources in a "reference house" which is defined as having a volume of 200 m^3 and an inner surface area of 350 m^2 with a ventilation rate of 0.5 h^{-1}. The relative indoor radon contributions attributable to various sources are given as follows:

building materials	80%	[first term in eq. (1.3)]
outside air	10%	[last two terms in eq. (1.3)]
water (toilets, showers, etc.)	5%	[contributors to second term in eq. (1.3)]
natural gas	4%	
liquified petroleum gas	< 1%	

It should be emphasized, however, that it is not clear what is contained in the "building materials" source category. The report [UN 80] does not specifically address whether this is only the contribution of radon from the building material itself, or whether it also includes transport of radon from soil gas through the building material.

Each of the terms in eq. (1.3) which contributes to the indoor radon concentration is associated with a form of transport. The first and last of these represent two physically different kinds of transport through building materials. The magnitude of the contribution of the last term is completely independent of the physical properties of the building materials, but depends instead on the dimensions of the crack(s) and the pressure difference between inside and outside. For this reason, such a form of transport is not considered in this report. Instead, this assessment is limited to the first transport source where the flux density, E, of radon atoms per second per square meter exhaling from a surface depends upon intrinsic physical properties of the material, such as effective diffusion coefficient, permeability, porosity, and radium-226 content.

Successive sections of this chapter include:

A. a description of the qualitative and quantitative characteristics of the physical model which forms the basis of our understanding of radon transport through building materials;

B. a critical review of previous work on the measurement of effective diffusion coefficients and permeabilities of building materials;

C. a brief discussion of some related transport problems which have been considered, or should be considered, in evaluating the source term, E, in eqs. (1.1) - (1.3) (e.g., transport through multilayered materials, or the effect of time variation of external parameters such as pressure or external radon concentration);

D. a summary of the time-dependent transport equations for radon in a porous medium, and a list of a set of solutions of both the steady-state and time-dependent transport equations for various transport cases; and

E. some conclusions and a recommendation that a series of measurements be made which are intended to determine a complete set of transport coefficients and related physical properties for several different building materials.

I.A. The Transport Model

I.A.1. Qualitative Description of Model

A building material is assumed to be a homogeneous porous medium, and transport of radon through the material is assumed to take place through its air-filled pore system. Transport of radon takes place in response to two different driving forces. A radon concentration difference is the driving force for diffusive transport through the air-filled pore system; and the associated transport coefficient is the effective diffusion coefficient of the material for radon. A second driving force is a pressure difference. Air in the pore system moves in response to a pressure difference and if radon is present, it moves with the air. The associated transport coefficient is the permeability coefficient of the material for air. In addition to these transport coefficients, which are unique for each building material, two other material constants which enter the transport equations for radon are the porosity (the ratio of pore volume to total volume), and the volume production rate of radon which is free to move through the pore system. It will be seen in the next section that there are very few measurements of the transport coefficients of radon in building materials. It should be recognized, however, that there is an extensive geological literature on radon migration in the ground which is comprehensively reviewed by Tanner [Tan 64; Tan 78]. Many of the qualitative aspects of radon migration in the ground, described in Tanner's reviews, are transferable to the discussion of transport of radon in building materials.

In addition to Tanner's reviews of the transport of radon in porous media, the monographs by Scheidegger [Sch 74], Beran [Ber 68], and Carman [Car 56], and the book edited by DeWiest [DeW 69] deal with aspects of the problem from different points of view. Diffusion or mass transport by fluid flow through porous or granular materials is influenced by pore shape, size, and volume, particle packing and size distribution (see, for example, Chapter II which describes the pore system in concrete). A hierarchy of parameters can be introduced to characterize a pore system [Sch 74; Ber 68; Car 56]. The simplest of these, and the only one which we will use explicitly, is the porosity which is defined as the ratio of the volume of the pore spaces in the material to the total volume of the material.

If the pore volume in a material such as concrete or soil is filled with a fluid such as air, diffusive transport of radon through the pore system takes place primarily via diffusion through the air-filled pores because the coefficient of diffusion of radon through air is at least 8 or 9 orders of magnitude greater than the coefficient of diffusion through the crystalline or glassy materials composing the matrix of the porous medium [Tan 64; Tan 78]. The influence of external variables, such as temperature and pressure on the one hand and humidity on the other, on the effective diffusion coefficient of radon in a porous material may be quite different. In the case of moderate temperature and pressure changes, the configuration of the pore system would not be expected to change. Thus, any variation of the effective diffusion coefficient with changes in these variables would simply reflect the weak dependence of the diffusion coefficient of radon in air (D) on these same variables (e.g., $D \sim T^{1/2}$ where T is the absolute temperature). On the other hand, when the external humidity is increased in the case of a material such as concrete (see Section II.D.), one would expect additional condensation of water in the finer capillaries of the pore system. Thus, although the dependence of the diffusion coefficient of radon in air on water content is weak, an alteration in the characteristics of the pore system in concrete with increasing water vapor pressure may signal a strong dependence on humidity of the effective diffusion coefficient of radon in concrete. Moisture content has a different effect which is not related to the transport of radon in the sense in which we have been using it. Tanner [Tan 64; Tan 78] has discussed the exhalation of radon into the pore system from trace amounts of radium in the matrix material. Viewed from the point of view of the pore system, there is a pore-volume production rate of radon which appears to be enhanced if the "humidity in the pores" is increased. [Tanner also mentions observations of exhalation of radon from the ground in the extreme case of saturation by a heavy rainfall. In such a case, there is a temporary decrease in exhalation which results from complete filling or blocking of the pore system near the surface by water (in which radon is less mobile)].

I.A.2. Quantitative Characterization of Model

As indicated, the proposed model for the transport of radon in or through a building material assumes that the building material is a porous medium and that transport of radon takes place through the air-filled pore system of the material. In response to a concentration difference of radon, there is diffusive transport of the radon in a direction which tends to equalize concentration. The empirical law which relates the diffusive current to a concentration difference is known as Fick's law. Given a thin slab of building material of thickness ΔZ and given fixed concentrations C_2 to the right of the slab and C_1 to the left, then the steady diffusive flux density, j_D, (net number of gas molecules passing from left to right through unit area of the slab per second) is proportional to the concentration difference $\Delta C = C_2 - C_1$, and inversely proportional to the thickness, ΔZ. The limiting form for small thicknesses and small concentration differences is a statement of Fick's law,

$$j_D = -D_e \left(\frac{dC}{dZ}\right) , \tag{1.4}$$

where D_e is the effective diffusion coefficient of the gas through the air in the pore system of the material.

Fick's law has been verified experimentally for diffusion of gases in homogeneous media such as gases, liquids, and crystalline or glassy solids. However, in a detailed study of diffusive transport through a given building material, it should be established experimentally rather than be assumed that there is an effective diffusion coefficient relating diffusive flux to concentration gradient in that material. Thus, if both the concentration difference, ΔC, and the thickness ΔZ are increased proportionally, the diffusive flux should be unaltered (except for a possible radioactive decay correction in the case of radon). It should be noted that if the validity of Fick's law is established for one or more simple gases in a given building material, then there is no reason to doubt that Fick's law would also hold for radon.

There is a similar operational procedure for establishing an empirical transport relation between air flow through a slab of porous material and pressure difference. The empirical law which relates the air volume current, v, (cm^3 per second per cm^2) through a porous material to the pressure difference, $\Delta P = P_2 - P_1$, maintained across a slab of thickness ΔZ is known as Darcy's law,

$$v = -\frac{k}{\eta}\frac{dP}{dZ} , \tag{1.5}$$

where η is the dynamic viscosity of the gas (air) in the pore system and k is the permeability coefficient of the porous material. Similar questions regarding the validity of Darcy's law for a building material arise as in the case of Fick's law. However, the validity of either law should imply the validity of the other.

I.B. Transport Coefficients for Building Materials

Very few measurements of the transport coefficients of radon in building materials have been made. A critical review and evaluation of this previous work follows.

I.B.1. Diffusion Coefficient

Culot, Olson, and Schiager [COS 76; COS 73] have determined an effective diffusion coefficient, D_e, for radon in a particular concrete of unmeasured porosity, ε. For a range of assumed values of the porosity (0.05-0.25), their value of the diffusion coefficient ranged from 1.69×10^{-5} to 3.08×10^{-5} $cm^2 \cdot s^{-1}$.

Jonassen and McLaughlin [JoM 76; JoM 78], in a more complete study of a particular concrete containing trace amounts of radium-226, determined values for the effective diffusion coefficient, porosity, and the production rate of radon. The values obtained for D_e and ε were 3.07×10^{-5} $cm^2 \cdot s^{-1}$ and 0.265, respectively. It should be noted that this determination [JoM 76; JoM 78] was an integral part of the measurement of the exhalation rate of the concrete. In the course of carrying out the exhalation measurements (by following the build-up of radon in a closed pressure chamber containing the concrete, see Section IV.B), Jonassen and McLaughlin also obtained indirect evidence for the validity of the assumed model for diffusion of radon in concrete. Namely, concrete appears to behave like an ideal porous solid for the diffusive transport of radon. This conclusion is based on the agreement between the observed radon concentration build-up with time and the form of the theoretically predicted build-up curve.

In the same report, Jonassen and McLaughlin [JoM 78] carried out the difficult experiment of measuring the steady-state concentration profile of radon across a 30-cm thick concrete wall. The measurements were made by drilling holes to different depths in the wall to insert sampling tubes through which air-radon samples could be collected in evacuated liter flasks. This admittedly crude experiment, which sampled pore air from an ill-defined volume surrounding the open end of each sampling tube, yielded a concentration profile similar in form to the theoretically predicted one (see Case III in Section I.D.3).

More recently, Stranden and Berteig [StB 80] have determined a diffusion coefficient and porosity for radon in concrete, brick, and LECA (a light expended clay aggregate). There are no other known measurements of diffusion coefficients of radon in concrete or any other building material such as phosphogypsum board.

I.B.2. Permeability Coefficient

Data for the permeability coefficient of air in concrete are available mainly because concrete structures such as sewage tanks, gas purifiers and containment vessels in nuclear reactors are required to be airtight under a specified internal pressure. For example, the steady-state flow of air through a certain concrete which is 10-cm thick is 1.45 x 10^{-6} $cm^3 \cdot s^{-1} \cdot cm^{-2}$ for a pressure difference of 26 mm Hg [Nev 71]. (The time to establish steady-state for concrete up to 23-cm thick is given as "several hours".) Assuming that the concrete contains trace amounts of radium-226, one can solve, in the steady-state, the combined transport equation for diffusion of radon and transport of radon by air flow under a pressure gradient across a 10-cm thick slab of concrete. If we assume the above stated flow velocity (i.e., 1.45 x 10^{-6} $cm^3 \cdot s^{-1} \cdot cm^{-2}$), an effective diffusion coefficient of $D_e = 3.1 \times 10^{-5}$ $cm^2 \cdot s^{-1}$, and a porosity of $\varepsilon = 0.1$, then we can calculate the ratio of the radon fluxes from the two surfaces of the slab of concrete (see Case V in Section I.D.5)*. This ratio is 1.16.

The only experimental investigation of the influence of a pressure change on the exhalation of radon from concrete known to us is by McLaughlin and Jonassen [McJ 78]. They sealed 60-cm diameter exhalation cans on a 30-cm thick concrete wall and maintained constant underpressures relative to atmospheric pressure in the range 5-25 mm Hg during the build-up of radon in the cans. They found that the exhalation rate at an underpressure of 25 mm Hg exceeded the zero underpressure or atmospheric pressure exhalation rate by a factor of 6. One difficulty with the interpretation of this measurement is that in order to prevent leaks, an epoxy resin sealant was applied to the wall surface surrounding the 60-cm diameter exhalation can to a distance of approximately 30 cm. The steady-state concentration of radon near the wall surface under the epoxy coating should be considerably enhanced, and the effect of this distortion in the concentration field on the exhalation rate measurement is difficult to estimate.

It should also be noted that for a 25 mm Hg underpressure, the observed total flow velocity into the exhalation can of 2 $L \cdot h^{-1}$ is equivalent to 2 x 10^{-4} $cm^3 \cdot s^{-1} \cdot cm^{-2}$. This flow velocity [McJ 78] is approximately 400 times greater than that for the thickness-scaled flow velocity of air through 30 centimeters of the concrete mentioned at the beginning of this section (1/3 x 1.45 x 10^{-6} $cm^3 \cdot s^{-1} \cdot cm^{-2}$) [Nev 71]. This large difference in permeability may reflect either a large difference in the permeability properties of the two concretes or the fact that the flow velocity, which must be measured under specified high-pressure difference conditions [Nev 71], does not scale linearly to the small pressure differences of interest here, or both. Any nonlinearity for large pressure differences would, most likely, lead to an underestimate of the flow velocity at low pressure differences. The resolution of the nature of the above difference requires further investigation. It is shown in Section I.D.5 that if the permeability of a concrete is as large as that indicated in the McLaughlin-Jonassen experiment [McJ 78], then the radon transport properties of a 10-cm slab should be dominated by the effect of a 25 mm Hg pressure difference.

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*This particular solution of the steady-state transport equation has not appeared in the literature.

I.C. Related Transport Problems

I.C.1. Transport Through Surface Sealants

In many circumstances, a building material in a basement floor or wall or a room wall has a multilayered or composite character. This composite nature may arise in the fabrication process, the esthetic addition of paint or other surface coverings, or the application of a surface sealant as a remedial measure to inhibit the transport of radon through concrete. There appear to be relatively few measurements of the diffusion coefficient of radon and the solubility coefficient of radon in a surface sealant [BCP 74; HEG 75]. The two examples where measurements have been made were for polycarbonate film [HEG 75] and for asphalt emulsion [BCP 74]. In this connection, there is an extensive literature devoted to the measurement of the product of the diffusion coefficient and the solubility for simple gases in polymeric films (see, for example, the review by Hwang, Choi, and Kammermeyer [HCK 74]. This product, which occurs naturally in the analysis of the experiments, has also been given the name "permeability". It should not be confused with the permeability k in Darcy's law.

In addition to the foregoing measurements, qualitative searches have been made [COS 73] to select from among those sealants which are available commercially the one(s) most effective in inhibiting the diffusive transport of radon through a 3-inch layer of concrete.

The modeling of diffusive transport through multilayer slabs is straightforward. A different class of more complex radon transport problems arises if flaws such as pinholes or cracks exist in one of the layers. Then the steady-state concentration field in the vicinity of the flaw is no longer one-dimensional. However, mathematical techniques analogous to those used in determining edge corrections in capacitor configurations are available for estimating the diffusive flux through the flaw. Other direct, brute-force numerical solution methods are also available [EdB 80].

I.C.2. Identification of the Dominant Transport Mechanism

There is no clear-cut answer to the question: Is the diffusion of radon through building materials the dominant transport mechanism contributing to the steady-state indoor concentration of radon? In part, this is because the dominant radon sources for a given building are very situational and site-specific depending on the radon concentration in the soil gas surrounding the building's foundation, the integrity of the building envelope, concentration of radon in the building's services (water, natural gas), etc. It is possible, however, to estimate the steady-state indoor radon concentration if diffusion is the only transport mechanism. This estimate requires knowing: 1) the ventilation rate of the indoor air; 2) the source strengths of radon (i.e., the production rate in the building material and the external concentrations of radon at the foundation wall and floors); and 3) the diffusive transport coefficients, porosities, and thicknesses of slabs of building materials (recall eq. (1.3)).

One example of such an estimate is a report to the Atomic Energy Control Board of Canada [DSM 78]. From measurements of external concentrations of radon and radon flux density, assumed source strength of radon in the concrete, diffusion coefficient and porosity estimates of Culot et al. [COS 76] for concrete, estimates of ventilation rates in 20 houses at Elliot Lake, and an average indoor radon concentration of 4 $pCi \cdot L^{-1}$, the two principal conclusions of the report were: first, that the dominant source of diffusing radon had to be in the concrete itself; and second, since the measured diffusive flux densities were insufficient to maintain the observed indoor radon concentration, that radon diffusion from or through concrete could not be a significant source of radon at Elliot Lake. This conclusion was based on a relatively high assumed value of the ventilation rate (1 air change per hour). However, if the assumptions are accepted, then the useful conclusion is reached that at Elliot Lake, radon must be entering the houses through cracks in basement walls, or holes through which utilities enter. The major source of radon in the buildings was not concrete or other building materials, but rather was soil gas containing radon which entered through discontinuities in the construction, cracks in floors and basement walls, holes through which utilities enter, drains with improper traps, etc. This may not be a typical situation for houses, however, since Elliot Lake is in a uranium district and may be geologically atypical.

In contradistinction, a twenty-house study in the New York-New Jersey area, performed by the Environmental Measurements Laboratory [GeB 78], showed that the indoor radon concentration could be accounted for reasonably well on the basis of two radon sources, the radon flux density through the basement floor and the radon concentration in the water supply, and an inferred average ventilation rate. Measurement data for these variables were subjected to multiple regression analyses, and the results were then used to obtain calculated values of the average annual radon concentrations which were in rough agreement with directly determined values. Although the observed mean indoor radon concentrations correlated reasonably well with radon exhalation rates from basements, this does not necessarily imply that there is a causal relationship, and it may only be an indication of the internal consistency in the radon concentration and radon flux measurement data. In this work [GeB 78], exhalation rates were measured over floor cracks in older houses as well as over intact concrete floors in newer houses. Within any given building, wide variations in radon exhalation were observed among simultaneous measurements and between successive sets of measurements. Although the radium-226 concentration in the soil around the houses is fairly uniform at approximately 1 $pCi \cdot g^{-1}$, and all building codes in the New York-New Jersey area require a 4-inch thick concrete floor in the basements, a 10-fold range in the average radon flux from the floors was found. This implies a correspondingly large range of radon input rates. The causes of this variation, part but not all of which can be explained by cracks, are unknown. It was suggested [CoM 80] that these wide variations, as well as similar 100-fold ones seen in houses with unvented crawl spaces, indicate that the rate depends on the type of soil. It was also pointed out that soil type is only one of many factors. Depending on meteorological conditions above the soil, 100-fold changes in a 12-hour period can be observed (see, for example, Chapter III). Soil is a major source in many cases, but it appears that this is not necessarily universal.

Most European investigators believe that the dominant source of indoor radon is from diffusion from building materials. In Denmark, for example, Jonassen and McLaughlin [JoM 76] concluded that radon originated primarily from building materials since indoor radon concentrations calculated from exhalation rates of walls and floors were in reasonable agreement with direct measurements of the average radon concentration. Significantly, no differences in the exhalation rates between floors and walls in contact with soils and walls not facing soils were found.

More recently, Goldsmith, et al. [GHA 80] investigated the routes of entry of radon into two buildings in the Canonsburg (Pennsylvania) Industrial Park which were built on low-level radium-contaminated ground. They identified and investigated a number of sources and pathways, and concluded that approximately 40-50% of the indoor radon concentration was due to diffusion through the intact concrete floors, and the remainder was from cracks, floor drains, and other openings in the building envelopes.

I.C.3. Time-Dependent Effects

Most of the investigations of the balance eq. (1.1) and the transport eq. (1.6) focus on steady-state behavior. It is possible to analyze these equations and determine effective relaxation times to establish steady-state conditions. For example, if the external radon concentration changes to a different steady value (in response to a change in meteorological conditions, or simply as a diurnal variation), then the relaxation time for establishing a new steady-state concentration profile in a 10-cm thick concrete slab is of the order of 3.5 days (see Case VI in Section I.D.6). In such a case, the effect of diurnal variations in radon concentration is "averaged out" and the transport of radon through the concrete slab will depend only on the time-averaged radon concentrations outside the slab. In a similar analysis of the relaxation of the pressure in the pores of the concrete following a change in the external pressure, it is shown in Case VII of Section I.D.7 that the relaxation time (for the same 10-cm thick concrete slab) to establish steady-state conditions is of the order of 1 hour.

I.D. Quantitative Discussion of Various Transport Cases

The simplest possible geometrical configuration in which to discuss the transport of radon in a porous medium will be considered. Namely, we consider one-dimensional transport through a slab where it is assumed that all concentrations and physical properties are constant in directions parallel to the surface of the slab and that all characteristic lengths in the normal direction are small compared to the transverse dimensions. Even with the restriction to one-dimensional configurations, there is a large number of interesting cases of steady-state transport which have been analyzed in different contexts and which are relevant to the discussion of radon transport in and through building materials. Before enumerating these illustrative cases, the equation for one-dimensional transport of a radioactive gas such as radon in a porous medium in the presence of a concentration gradient and a pressure gradient will be presented without derivation. The equations are contained in Clements and Wilkening [ClW 74] where some earlier references may be found. A derivation is given in Clements' thesis [Cle 74] which is similar to the derivation of Bulashevich and Khairitdinov [Buk 59]. Other references to the older literature may be found in Tanner [Tan 64; Tan 78]. Additional discussion and references to the literature on the transport equation are given by Culot et al. [COS 76]. This last reference is particularly useful in pointing to confusion in the literature regarding values quoted for the effective diffusion coefficient D_e.

The following definitions are used throughout this section, and the boundary conditions will be stated for each case:

ε porosity, ratio of pore volume to bulk volume

k permeability coefficient in expression for Darcy's law, m^2

η dynamic viscosity of fluid in pores, $kg \cdot m^{-1} \cdot s^{-1}$

ρ density of fluid in pores, $kg \cdot m^{-3}$

D_e effective diffusion coefficient in porous medium, coefficient in Fick's law, $m^2 \cdot s^{-1}$

C	^{222}Rn concentration in the pore system, in units of radon activity per unit volume of pore system, $\mathrm{pCi \cdot m^{-3}}$
Z	distance measured in direction of transport, m
λ	decay constant of ^{222}Rn, $\lambda = (\ln 2)/T = 2.1 \times 10^{-6}\ \mathrm{s^{-1}}$ where T = ^{222}Rn halflife
ϕ	^{222}Rn production rate, number of ^{222}Rn atoms produced per unit time per unit pore volume that are free to migrate through the pore system (i.e., ^{222}Rn produced by the effective ^{226}Ra), $\mathrm{pCi \cdot s^{-1} \cdot m^{-3}}$
ℓ	diffusion length, or lifetime diffusion-distance, a convenient length scale characterizing diffusion of atoms which undergo radioactive decay, $\ell = (D_e/\varepsilon\lambda)^{1/2}$,
v	fluid volume current density, volume of fluid in pore system flowing per unit time per unit geometric area, $\mathrm{m^3 \cdot s^{-1} \cdot m^{-2} = m \cdot s^{-1}}$; for pressure-induced flow, according to Darcy's law $v = - \frac{k}{\eta} \frac{\partial P}{\partial Z}$ where P is the pressure, $\mathrm{kg \cdot m^{-1} \cdot s^{-2}}$.
j_D	diffusion current density or flux density of radon, radon activity per unit time per unit geometric area transported by molecular diffusion through the pore system, depends on the concentration gradient through Fick's law, $\mathrm{pCi \cdot s^{-1} \cdot m^{-2}}$, $j_D = - D_e \frac{\partial C}{\partial Z}$
j_T	transport current density of radon, number of radon atoms per unit time per unit geometric area transported by fluid flow, $\mathrm{pCi \cdot s^{-1} \cdot m^{-2}}$, $j_T = vC$
P_o, p	the pressure P is assumed to be the sum of a steady component, P_o, and a small time varying component, p, in deriving eq. (1.8) using Darcy's law.

The time-dependent one-dimensional transport equations can be written as:

$$\frac{\partial C}{\partial t} = \frac{D_e}{\varepsilon} \frac{\partial^2 C}{\partial Z^2} - \frac{1}{\varepsilon} \frac{\partial (vC)}{\partial Z} - \lambda C + \phi \tag{1.6}$$

$$v = - \frac{k}{\eta} \frac{\partial P}{\partial Z} , \quad \text{(Darcy's law)} . \tag{1.7}$$

In the case where the effect of small atmospheric pressure variations, p, are being considered, the variation of pressure in the porous medium is governed by the equation [ClW 74; Cle 74; Fuk 55]

$$\frac{\partial^2 p}{\partial Z^2} = \frac{\varepsilon\eta}{kP_o} \frac{\partial p}{\partial t} . \tag{1.8}$$

Most of the discussion to follow will deal with the steady-state condition, in which case eq. (1.6) becomes

$$\frac{D_e}{\varepsilon}\,\frac{d^2C}{dZ^2} - \frac{1}{\varepsilon}\,\frac{d(vC)}{dZ} - \lambda C + \phi = 0\ . \qquad (1.9)$$

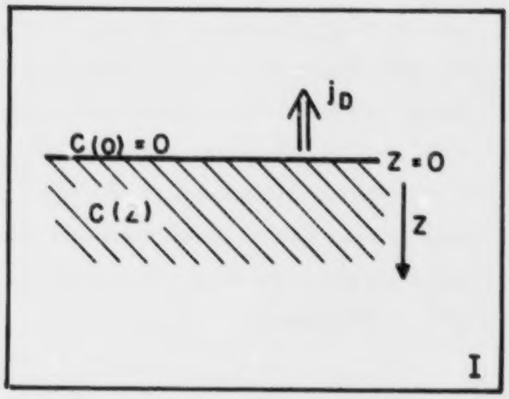
A few remarks regarding the material parameters may be useful. For transport of radon in unsaturated (with water) materials, the fluid filling the pore system is air, so the values of ρ,η (the density and viscosity) are those for air. The material constants of the building material (porous medium) are: ε, k, D_e, and ϕ. There is no simple relation between the diffusion coefficient of radon in air and the effective diffusion coefficient of radon through the air-filled pore system of a porous material such as concrete. The porosity of concrete is a measure of the volume fraction of the air-pore system. However, the average number of pores which connect opposite faces of a concrete slab and the average length of such pores effectively reduce the diffusion coefficient of radon in air by four orders of magnitude.

The first few cases are intended to familiarize the reader with the notion of exhalation of radon from a (building) material, and to emphasize that the radon flux density from a surface is not an intrinsic property of a material but can depend on the boundary condition at another surface. Until stated otherwise in these cases, the air volume current density is assumed to be $v = 0$.

I.D.1. Case I: Exhalation from the Ground into Open Air

This case considers:

The concentration profile of radon in the soil and radon flux density from the ground into the open air assuming that the soil contains a uniform distribution of radium-226 giving rise to a volume production rate ϕ which is constant everywhere below the soil-air interface, and further assuming that the concentration of radon at the open soil-air interface is maintained at $C = 0$ by natural circulation of the air.



The concentration profile of radon in the soil is then

$$C(Z) = \frac{\phi}{\lambda}\left\{1 - \exp(Z/\ell)\right\}, \qquad Z \leq 0 \tag{1.10}$$

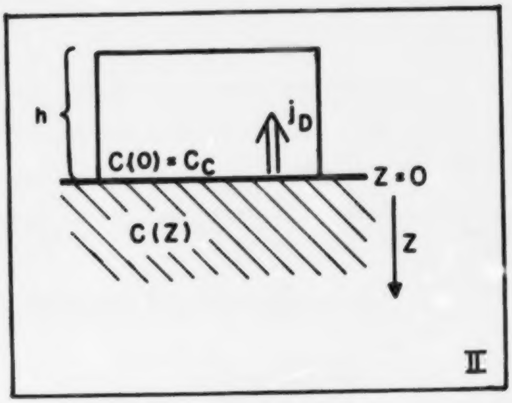
where C(Z) ranges from 0 at $Z = 0$ to ϕ/λ deep in the soil. The characteristic length, $\ell = (D_e/\varepsilon\lambda)^{1/2}$, a diffusion length or lifetime-diffusion distance, is a measure of the distance in which there is an e-fold variation in radon concentration near the soil surface. The diffusion current density of radon at the soil surface (i.e., the radon flux density in $pCi\cdot s^{-1}\cdot m^{-2}$) is

$$j_D = - D_e \left.\frac{\partial C}{\partial Z}\right|_{Z=0} = \phi(D_e\varepsilon/\lambda)^{1/2} = \phi\varepsilon\ell . \tag{1.11}$$

I.D.2. Case II: Exhalation from the Ground into a Closed Space

This case considers:

> the concentration profile of radon in the soil and radon flux density from the ground into a closed collecting container assuming all other conditions are identical to Case I.



This case is similar to Case I except that it models the presence of a collecting container which is used for in situ measurements of radon flux density by the accumulation method (see Section IV.C.1.). For these measurements, the accumulator (typically a cylindrical can closed on top and open on the bottom) is placed in close contact with the ground. If the diameter of the can is made sufficiently large, then the steady-state diffusion field underneath the can will be one-dimensional. In this case, if it is further assumed that the air in the can is well-mixed so that no concentration gradients of radon develop, then the steady-state diffusion flux density from the soil-surface into the can will just compensate the volume decay rate of radon in the can. The appropriate boundary condition at the soil-air interface is

$$- A D_e \left.\frac{\partial C}{\partial Z}\right|_{Z=0} = Ah\, C_c\lambda \tag{1.12}$$

where A = cross-sectional area of the can, h = height of the can, C_c = concentration of radon in the can as well as the limiting value of C(Z) at the soil-air interface. The concentration profile of radon in the soil in this case is

$$C(Z) = \frac{\phi}{\lambda}\left\{1 - \frac{\exp(Z/\ell)}{1+\varepsilon\ell/h}\right\}, \qquad Z \leq 0 \tag{1.13}$$

and the flux density of radon into the can (i.e., the constrained areal exhalation rate) is

$$j_D = \varepsilon\phi\ell[1+\varepsilon\ell h^{-1}]^{-1} \tag{1.14}$$

which is just the free areal exhalation rate in Case I [eq. (1.11)] reduced by the factor $[1+\varepsilon\ell h^{-1}]^{-1}$. The concentration of radon in the collecting can is

$$C_c = C(0) = \frac{\phi}{\lambda}\left(\frac{\varepsilon\ell}{\varepsilon\ell+h}\right).$$

or

$$C_c = C(-\infty)\left(\frac{\varepsilon\ell}{\varepsilon\ell+h}\right)$$

where the factor $\varepsilon\ell/(\varepsilon\ell+h)$ is the ratio of two volumes, the volume of the pore space under the container to a depth of one diffusion length to the sum of that volume and that of the container. In the limit in which the height of the collecting can is small compared to the diffusion length ℓ, the concentration profile C(Z) in eq. (1.13) is simply

$$C(Z) \cong \phi/\lambda$$

and the flux density into the can, eq. (1.14), is

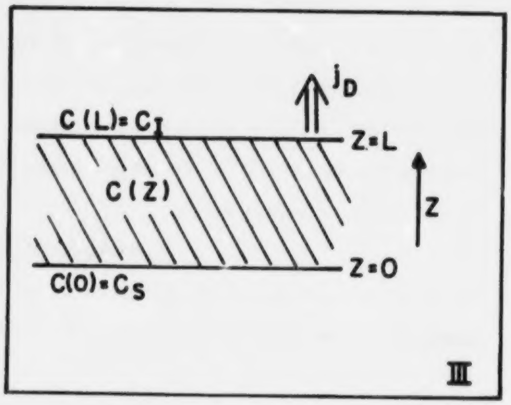
$$j_D \cong h\phi\ .$$

In this connection, two remarks may be made: 1) In the limit in which the height of the can approaches zero, the boundary condition [eq. (1.12)] becomes $\left.\frac{\partial C}{\partial Z}\right|_{Z=0} = 0$. This boundary condition is appropriate at a surface which is impermeable to the diffusing gas and leads to the constant concentration profile, $C(Z) = \phi/\lambda$. The production rate of radon, ϕ, is balanced by the decay rate, λC. 2) The concept of free and constrained exhalation rates has also been used by Jonassen and McLaughlin [JoM 78] in an experimental determination of exhalation properties of building materials. Some of their modeling results are summarized in Case III. Jonassen and McLaughlin also attribute the use of eq. (1.9) (with v=0) to Krisiuk et al. [KTS 71].

I.D.3. Case III: Exhalation from One Side of a Slab

This case considers:

> the concentration profile of radon and radon flux density from a slab of thickness L containing a uniform concentration of radium-226 and, in addition, the radon concentrations above and below the slab are maintained at C_I and C_s, respectively.



This configuration has been considered in an Atomic Energy Control Board of Canada Report [DSM 78] to determine the principal sources of indoor radon in a survey of 20 houses in Elliot Lake. Other special cases of this configuration have been treated by Culot et al. [COS 76], Jonassen and McLaughlin [JoM 78], and Clements [Cle 74]. The concentration profile in this case is

$$C(Z) = \frac{\phi}{\lambda}\left[1 - \frac{\sinh(Z/\ell) + \sinh[(L-Z)/\ell]}{\sinh(L/\ell)}\right] + C_I\,\frac{\sinh(Z/\ell)}{\sinh(L/\ell)} + C_S\,\frac{\sinh[(L-Z)/\ell]}{\sinh(L/\ell)} \qquad (1.15)$$

for $0 \leq Z \leq L$. This solution of the steady-state eq. (1.9) satisfies the boundary conditions $C(0) = C_s$ and $C(L) = C_I$ below and above the slab, respectively. The flux density of radon across the surface of the slab at $Z = L$ is

$$\begin{aligned} j_D &= -\,D_e\,\frac{\partial C}{\partial Z}\bigg|_{Z=L} \\ &= (\varepsilon\lambda D_e)^{1/2}\left[\frac{\phi}{\lambda}\,\frac{\cosh(L/\ell)-1}{\sinh(L/\ell)} + \frac{C_s}{\sinh(L/\ell)} - C_I\,\frac{\cosh(L/\ell)}{\sinh(L/\ell)}\right]. \end{aligned} \qquad (1.16)$$

If we identify ϕ with the radon production rate in a slab of concrete, C_s with the radon concentration of soil gas in contact with the outside surface of the slab and C_I with the inside radon concentration, it can be seen in eq. (1.16) that these parameters enter separate, independent contributions to the flux density of radon at the interior surface of the slab. The relative contributions to j_D in eq. (1.16) from "typical" values of ϕ and C_s are displayed graphically in [DSM 78] for a range of thicknesses of concrete ($5\text{cm} \leq L \leq 20\text{cm}$), and a range of typical values of the effective diffusion coefficient of concrete [COS 76] ($1.7 \times 10^{-5}\ \text{cm}^2\cdot\text{s}^{-1} < D_e < 3.1 \times 10^{-5}\ \text{cm}^2\cdot\text{s}^{-1}$) and porosity of concrete ($0.05 < \varepsilon < 0.25$). The diffusion length $\ell = 10$ cm for $D_e = 2.1 \times 10^{-5}\ \text{cm}^2\cdot\text{s}^{-1}$, $\lambda = 2.1 \times 10^{-6}\ \text{s}^{-1}$ for radon-222, and $\varepsilon = 0.1$. If the thickness, L, is appreciably larger than the diffusion length, ℓ, then the flux density j_D in eq. (1.16) can be written approximately as

$$\begin{aligned} j_D &\cong (\varepsilon\lambda D_e)^{1/2}\left[\frac{\phi}{\lambda}\,(1-2e^{-L/\ell}) + 2C_s e^{-L/\ell} - C_I(1 + 2e^{-2L/\ell})\right] \\ &\cong (\varepsilon\lambda D_e)^{1/2}\left[\frac{\phi}{\lambda} + 2C_s e^{-L/\ell} - C_I\right], \qquad L \gg \ell\ . \end{aligned} \qquad (1.17)$$

The first term in eq. (1.17) is identical with the expression for the exhalation of radon from the ground into the open air in Case I, eq. (1.11). The negative contribution, $-C_I$, in eqs. (1.16) and (1.17) represents a back-diffusion contribution to the flux density j_D from the radon above the slab. The contribution of the external radon concentration C_s to the flux density, j_D, in eq. (1.17) is attenuated by the factor $e^{-L/\ell}$. If the thickness, L, is appreciably less than ℓ, then the flux density, j_D, in eq. (1.16) is approximately

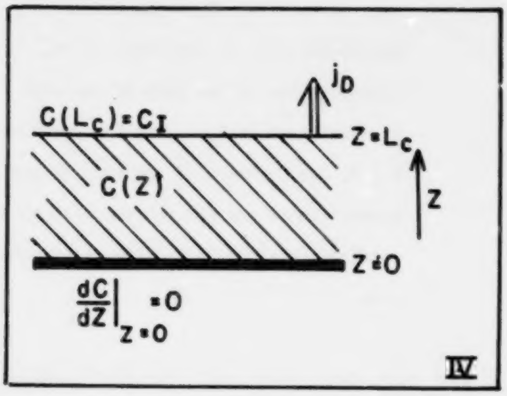
$$j_D \cong \varepsilon\phi L/2 + D_e\,(C_s - C_I)/L, \qquad L \ll \ell\ . \qquad (1.18)$$

The first term in eq. (1.18) is simply 1/2 of the volumetric rate of radon production corrected to geometric volume since ϕ is defined on the basis of pore volume. (The other 1/2 is the flux density in the opposite direction.) The second term in eq. (1.18) is simply the definition of the diffusive flux density in response to the concentration difference $C_s - C_I$ across the slab of thickness, L.

I.D.4. Case IV: Exhalation from a Slab with an Impervious Coating Applied to One Side

This case considers:

the radon concentration profile inside, and radon flux density from a slab containing a uniform distribution of radium-226 which is coated on one side with a material impervious to the transport of radon. In addition, there is a concentration C_I of radon in the air in contact with the uncoated side of the slab.



In this case, we are interested in calculating the surface flux density into a room from a layer or slab of building material containing radium-226 which is sealed on the external side (examples are a waterproof coating on a concrete surface or a vapor barrier on phosphogypsum board). The concentration profile in this case can be derived from that in Case III if the boundary concentration C_s in eq. (1.15) is set equal to C_I. For equal external concentrations, the concentration profile is symmetric about the midplane of the slab at $Z = L/2$. Therefore, the flux density across the mid-plane is zero and the concentration profile in the upper half of the slab (from $Z = L/2$ to $Z = L$) is the same as the profile for a slab of thickness $L/2$ with an impermeable layer at $Z = L/2$. The flux density through the exposed surface is, according to eq. (1.16),

$$j_D = (\varepsilon\lambda D_e)^{1/2}\left(\frac{\phi}{\lambda} - C_I\right)\frac{\cosh(L/\ell) - 1}{\sinh(L/\ell)} \qquad (1.19)$$

If we recognize that L in eq. (1.19) is equal to $2L_c$ where L_c is the thickness of the coated slab, we can rewrite eq. (1.19) as

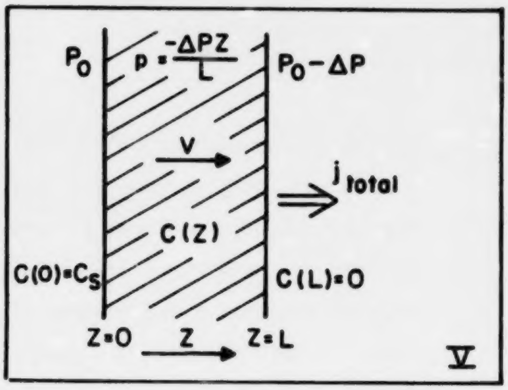
$$j_D = (\varepsilon\lambda D_e)^{1/2}\left(\frac{\phi}{\lambda} - C_I\right)\tanh(L_c/\ell) . \qquad (1.20)$$

Jonassen and McLaughlin [JoM 78] treated a configuration analogous to Case II, namely exhalation of radon from a bottom-surface coated slab into a closed space.

I.D.5. Case V: Influence of Darcy Flow on Exhalation from a Slab

This case considers:

the radon concentration profile and radon flux density from a slab containing a uniform distribution of radium-226 and, in addition; 1) the concentration to the left of the slab is maintained at C_s and to the right at $C = 0$, and 2) there is a steady forced permeation of air through the slab by a constant pressure difference across the slab.



In this case, the steady-state transport equation is, from eq. (1.9),

$$\frac{D_e}{\varepsilon}\frac{d^2C}{dZ^2} - \frac{v}{\varepsilon}\frac{dC}{dZ} - \lambda C + \phi = 0, \qquad 0 \le Z \le L \, , \tag{1.21}$$

where v > 0 corresponds to flow from left to right and v < 0 corresponds to the reverse. The steady-state concentration profile is

$$C(z) = \frac{\phi}{\lambda}\left\{1 - \frac{\exp[vZ/2D_e]\sinh[(L-Z)/\Lambda] + \exp[-v(L-Z)/2D_e]\,\sinh[Z/\Lambda]}{\sinh[L/\Lambda]}\right\} \tag{1.22}$$

$$+ C_s \exp[vZ/2D_e]\,\frac{\sinh[(L-Z)/\Lambda]}{\sinh[L/\Lambda]}$$

where $\Lambda^{-1} = \left[\frac{v^2}{4D_e^{\,2}} + \frac{\lambda\varepsilon}{D_e}\right]^{1/2} = \left[\frac{v^2}{4D_e^{\,2}} + \ell^{-2}\right]^{1/2}$. The length, Λ, is the diffusion length, ℓ, modified by the superimposed flow velocity, v. The total flow of radon per unit area through the surface at Z=L is

$$j_{total} = \left(-\,D_e\frac{dC}{dZ} + vC\right)\Bigg|_{Z=L}$$

$$= \frac{\phi}{\lambda}\left\{\frac{v}{2} + \frac{D_e}{\Lambda}\;\frac{\cosh(L/\Lambda) - \exp(vL/2D_e)}{\sinh(L/\Lambda)}\right\} + C_s\,\frac{D_e}{\Lambda}\;\frac{\exp(vL/2D_e)}{\sinh(L/\Lambda)} \tag{1.23}$$

There are two simple illustrative comparisons which can be made with the expression for current density, j_{total}, in eq. (1.23) at the surface Z = L. If there is no production of radon, the ratio of j_{total} for velocity v in the positive direction to j_{total} when v is reversed is

$$\frac{j_{total}(v > 0)}{j_{total}(v < 0)} = \exp[|v|L/D_e] , \qquad \phi = 0 . \tag{1.24a}$$

If $j_{total}(v)$ is compared to the radon flux density when there is no impressed flow velocity, $j_{total}(0)$, the corresponding ratio is

$$\frac{j_{total}(v)}{j_{total}(0)} = \exp(vL/2D_e) \left(\frac{\ell}{\Lambda}\right) \frac{\sinh(L/\ell)}{\sinh(L/\Lambda)} , \quad \phi=0 . \tag{1.24b}$$

If the external concentrations of radon on both sides of the slab are maintained at zero, then the corresponding ratios of the flux densities of radon through the surface at Z=L obtained from eq. (1.23) are

$$\frac{j_{total}(v>0)}{j_{total}(v<0)} = \frac{\frac{1}{\Lambda}\left(\frac{\cosh(L/\Lambda) - \exp(|v|L/2D_e)}{\sinh(L/\Lambda)}\right) + \frac{|v|}{2D_e}}{\frac{1}{\Lambda}\left(\frac{\cosh(L/\Lambda) - \exp(-|v|L/2D_e)}{\sinh(L/\Lambda)}\right) - \frac{|v|}{2D_e}} , \quad C_s=0 \tag{1.25a}$$

and

$$\frac{j_{total}(v)}{j_{total}(0)} = \frac{\frac{1}{\Lambda}\left(\frac{\cosh(L/\Lambda) - \exp(vL/2D_e)}{\sinh(L/\Lambda)}\right) + \frac{v}{2D_e}}{\frac{1}{\ell}\left(\frac{\cosh(L/\ell) - 1}{\sinh(L/\ell)}\right)} , \quad C_s = 0 . \tag{1.25b}$$

The air volume current density for a particular concrete [Nev 71] is $v = 1.45 \times 10^{-6}$ $cm^3 \cdot s^{-1} \cdot cm^{-2}$ when the pressure difference is 26 mm Hg and the thickness of the concrete is 10 cm. If we further assume that $D_e = 3.1 \times 10^{-5}$ $cm^2 \cdot s^{-1}$ and $\varepsilon = 0.1$, then the values of the ratios in eqs. (1.24a), (1.24b), (1.25a) and (1.25b) are, respectively, 1.60, 1.25, 1.16 and 1.08. Because of the symmetry of the special case of eq. (1.24a), that expression also is the ratio of the exhalations from the two surfaces (at Z=L and Z=0) when a flow velocity v is imposed from left to right. Using the foregoing permeability data in Darcy's law, one can obtain the relation $|v|L = 5.58 \times 10^{-7}|\Delta P|$ where the units on each side are ($cm^2 \cdot s^{-1}$), but ΔP is expressed in mm Hg. Substituting this expression for $|v|L$ in eq. (1.24a), one obtains the alternate form,

$$\frac{j_{total}(v > 0)}{j_{total}(v < 0)} = \exp\,(.018|\Delta P|) \tag{1.24c}$$

It can be seen from the values we have obtained for the ratios in eqs. (1.24b) and (1.25b) that imposing a pressure difference of 26 mm Hg increases the exhalation of radon by 25 and 8 percent, respectively. These increases follow by combining the permeability data of Neville [Nev 71] with the diffusion coefficient of 3.1×10^{-5} $cm^2 \cdot s^{-1}$ and porosity of 0.1. The flow-modified diffusion length, Λ, in the present case where $v = 1.45 \times 10^{-6}$ $cm \cdot s^{-1}$ is

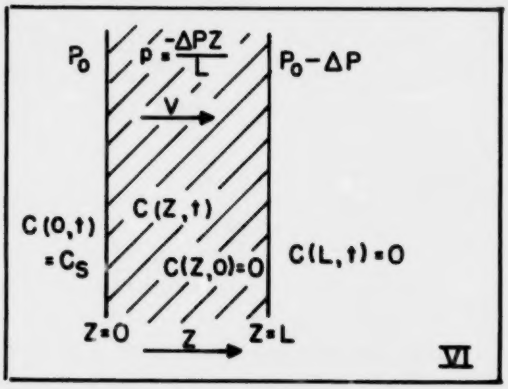
$$\Lambda = .962\ \ell$$

where the diffusion length $\ell = (D_e/\varepsilon\lambda)^{1/2} = 12.15$ cm. We noted in Section 1.B.2 that McLaughlin and Jonassen [McJ 78] obtained a permeability (and therefore a flow velocity) approximately 400 times greater than that quoted by Neville. The flow-modified diffusion length Λ is completely dominated by such a large velocity. Thus for $v = 5.8 \times 10^{-4}$ $\text{cm}\cdot\text{s}^{-1}$, the value of Λ is

$$\Lambda = 8.80 \times 10^{-3}\,\ell;$$

and the value of the ratio $j_{total}(5.8 \times 10^{-4})/j_{total}(0)$ in eq. (1.24b) is 208.2. For concrete with such a large permeability, pressure effects clearly dominate diffusion effects.

I.D.6. Case VI: Time Development of Steady-State Concentration Profile in a Slab



This case considers:

> the time development of the steady-state radon concentration profile in Case V when $\phi = 0$ and the initial concentration in the slab is $C(Z,0) = 0$.

In this case, the time-dependent transport equation is

$$\frac{\partial C(Z,t)}{\partial t} = \frac{D_e}{\varepsilon}\,\frac{\partial^2 C(Z,t)}{\partial Z^2} - \frac{v}{\varepsilon}\,\frac{\partial C(Z,t)}{\partial Z} - \lambda C(Z,t), \qquad 0 \le Z \le L \tag{1.26}$$

with $C(Z,0) = 0$, $C(0,t) = C_s$, and $C(L,t) = 0$. The solution to this initial value problem is [CaJ 59]

$$C(Z,t) = C_s \exp\left(\frac{vZ}{2D_e}\right)\left\{\frac{\sinh[(L-Z)/\Lambda]}{\sinh(L/\Lambda)} - \frac{2\pi}{L^2}\sum_{n=1}^{\infty}\frac{n\sin(n\pi Z/L)}{[\Lambda^{-2}+n^2\pi^2/L^2]}\exp[-(\Lambda^{-2}+n^2\pi^2/L^2)D_e t]\right\}. \tag{1.27}$$

The first term on the right in eq. (1.27) is the steady-state concentration profile already obtained in Case V, eq. (1.22) (with $\phi = 0$). If, in addition, the velocity v is set equal to zero in eq. (1.27), then the steady-state concentration profile in Case III, eq. (1.15) is recovered (with $\phi = 0$ and $C_I = 0$). Thus the solution of eq. (1.27) also includes the special case of the time development in Case III.

If the time, t, is sufficiently large compared to

$$t_R = [D_e(\Lambda^{-2}+\pi^2/L^2)]^{-1} = [D_e(\tfrac{1}{4}v^2 D_e^{-2} + \ell^{-2} + \pi^2 L^{-2})]^{-1},$$

then the time-dependent sum in eq. (1.27) may be neglected. Thus t_R is a measure of the "relaxation time" to the steady-state concentration profile (which includes both Cases III and V). In Case III where $v = 0$ and for $D_e = 3.1 \times 10^{-5}\ cm^2 \cdot s^{-1}$, $\varepsilon = 0.1$, and $\lambda = 2.10 \times 10^{-6}\ s^{-1}$, the "relaxation time" is

$$t_R = [2.10 \times 10^{-7} + 3.06 \times 10^{-4}\ L^{-2}]^{-1}, \quad \text{seconds}$$

where L is measured in cm. Thus for L = 10 cm, $t_R = 3.06 \times 10^5$ s (3.54 days). In Case V where we used the value $v = 1.45 \times 10^{-6}\ cm^3 \cdot s^{-1} \cdot cm^{-2}$ and the same values of D_e and ε, t_R is not significantly different from the value obtaired above

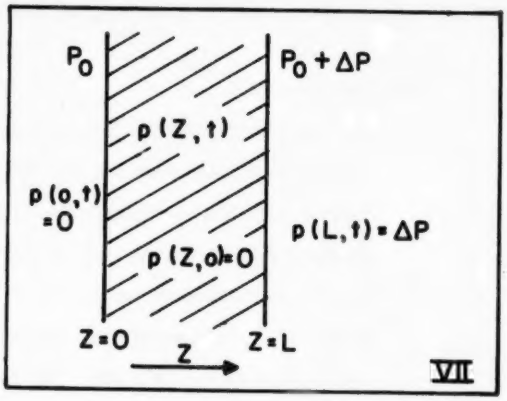
$$t_R = [2.27 \times 10^{-7} + 3.06 \times 10^{-4}\ L^{-2}]^{-1}, \quad \text{seconds.}$$

It appears from these numerical estimates that diurnal variations in radon concentrations outside a concrete slab will be averaged out because of the relatively large value of $t_R \sim 3.5$ d; and so transport of radon through the concrete will depend only on the time-averaged (over 3.5 days) values of radon concentrations outside the slab.

I.D.7. Case VII: Time Development of Steady-State Pressure Profile in a Slab

This case considers

the time-dependent pressure profile inside a slab of thickness L when the initial pressure inside the pore system and to the left of the slab is P_o, and at time t = 0 the pressure on the right is increased suddenly to $P_o + \Delta P$.



In this case, at any time t > 0, the pressure inside the slab is $P_o + p(Z,t)$. The transport equation for the time-varying component of the pressure inside the porous slab is given by eq. (1.8)

$$\frac{\partial p}{\partial t} = \frac{kP_o}{\varepsilon\eta} \frac{\partial^2 p}{\partial Z^2}, \tag{1.8}$$

an equation which is formally identical to the diffusion equation. The solution of the above initial value problem for eq. (1.8) is [Cra 75]

$$p(Z,t) = \frac{Z}{L}\Delta P - \frac{2}{\pi}\Delta P \sum_{n=1}^{\infty} (-1)^{n+1} \frac{1}{n} \sin\frac{n\pi Z}{L} \exp\left(-\frac{Dn^2\pi^2}{L^2}t\right) \tag{1.28}$$

where $D = \frac{kP_0}{\varepsilon\eta_0}$. The first term on the right side in eq. (1.28) is the steady-state pressure profile, a linear increase in pressure from P_0 to $(P_0 + \Delta P)$ across the slab. If the time t is large compared to $L^2/D\pi^2$, then every term in the time-dependent sum in eq. (1.28) may be neglected. Thus, $L^2/D\pi^2$ is a rough measure of the "relaxation time" for approach to the steady-state following a sudden change in pressure. Using Darcy's law to evaluate k/η from the permeability data cited in Case VI [Nev 71], one obtains a value for the relaxation time (in seconds):

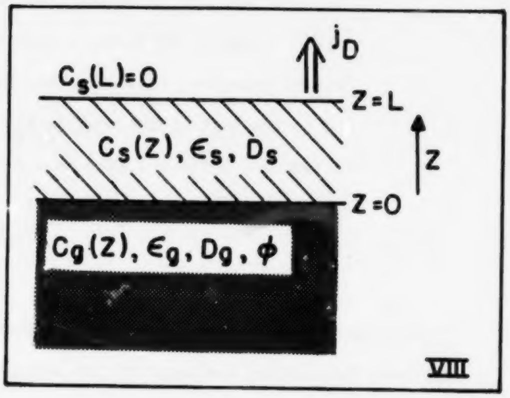
$$L^2/D\pi^2 = 24\ L^2, \quad \text{seconds} ,$$

where L is the slab thickness in cm. Thus the time required for the establishment of the steady-state pressure distribution in the pore system is of the order of one hour for 10 cm-thick concrete. It appears that, in contrast to the results in Case VI, the relaxation time in the present case is relatively short. This result will be useful in solving the time-dependent transport eq. (1.26) because the coefficient v can be assumed to adjust instantaneously to changes in pressure and maintain a steady-state profile.

I.D.8 Case VIII: Exhalation from a Slab Resting on the Ground

This case considers:

the radon concentration profile and radon flux density from the top surface of a slab of thickness L resting on the ground in the case where the soil beneath the slab contains a uniform distribution of radium-226, but the slab does not contain radium, and where the concentration of radon in the air above the slab is maintained at C = 0.



This configuration can be used to estimate the reduction in the flux density of radon from the ground which results when a concrete slab is interposed. The steady-state concentration profiles in the slab and in the ground are governed by the equations

$$\frac{D_g}{\varepsilon_g}\left(\frac{d^2C_g}{dZ^2}\right) - \lambda C_g + \phi = 0 \quad , \quad Z \leq 0 \tag{1.29}$$

and

$$\frac{D_s}{\varepsilon_s}\left(\frac{d^2C_s}{d\,Z^2}\right) - \lambda C_s = 0 \quad , \quad 0 \leq Z \leq L \tag{1.30}$$

The solutions of eqs. (1.29) and (1.30) must satisfy the following boundary conditions:

$$\text{at } Z = L : \quad C_s(L) = 0 \tag{1.31}$$

$$\text{at } Z = 0 : \quad C_s(0) = C_g(0) \tag{1.32}$$

$$-D_s \frac{dC_s}{dZ}\bigg|_{Z=0} = -D_g \frac{dC_g}{dZ}\bigg|_{Z=0} \tag{1.33}$$

$$\text{at } Z = -\infty: \quad C_g(-\infty) = \phi/\lambda \tag{1.34}$$

The conditions (1.32) and (1.33) at the ground-slab interface express the requirement that the radon concentration and flux density vary continuously there. The concentration profile in the ground is (for $Z \leq 0$)

$$C_g(Z) = \frac{\phi}{\lambda}\left[1 - \frac{\exp(Z/\ell_g)}{1 + \left(\frac{\ell_s}{\ell_g}\right)\left(\frac{D_g}{D_s}\right)\tanh\left(\frac{L}{\ell_s}\right)}\right] \tag{1.35}$$

and the concentration profile in the slab is

$$C_s(Z) = \frac{\phi}{\lambda}\left(\frac{\ell_s}{\ell_g}\right)\left(\frac{D_g}{D_s}\right)\left[\frac{-\sinh(Z/\ell_s) + \cosh(Z/\ell_s)\tanh(L/\ell_s)}{1 + \left(\frac{\ell_s}{\ell_g}\right)\left(\frac{D_g}{D_s}\right)\tanh\left(\frac{L}{\ell_s}\right)}\right] \tag{1.36}$$

The flux density of radon through the top surface of the slab is

$$\begin{aligned} J_D &= -D_s \frac{dC_s}{dZ}\bigg|_{Z = L} \\ &= \phi\, \varepsilon_s \ell_s \left\{\cosh\left(\frac{L}{\ell_s}\right)\left[1 + \left(\frac{\ell_s}{\ell_g}\right)\left(\frac{D_g}{D_s}\right)\tanh\left(\frac{L}{\ell_s}\right)\right]\right\}^{-1} . \end{aligned} \tag{1.37}$$

According to Case I, the flux density of radon through the bare ground-air interface is $\phi\varepsilon_s\ell_s$, i.e., the coefficient on the right-hand side of eq. (1.37). Thus, the ratio of the flux density of radon through the soil-air interface in the absence of the slab to the flux density through the top surface of the slab is

$$\frac{\phi\varepsilon_s\ell_s}{J_D} = \left[\cosh\left(\frac{L}{\ell_s}\right) + \left(\frac{\ell_s}{\ell_g}\right)\left(\frac{D_g}{D_s}\right)\sinh\left(\frac{L}{\ell_s}\right)\right] \tag{1.38}$$

If the slab thickness, L, is large compared to the slab diffusion length, ℓ_s, then the approximate relation obtained from eq. (1.38) is

$$\frac{\phi\varepsilon_s\ell_s}{J_D} \cong \frac{1}{2}\left[1 + \left(\frac{\ell_s}{\ell_g}\right)\left(\frac{D_g}{D_s}\right)\right]\exp\left(\frac{L}{\ell_s}\right) \quad , \quad L \gg \ell_s \quad . \tag{1.39}$$

For illustrative purposes, consider the reduction of the radon flux density from soil by the interposition of a concrete slab of thickness L = 10 cm, with a diffusion length ℓ_s = 10 cm and diffusion coefficient $D_s = 2 \times 10^{-5}\ cm^2 \cdot s^{-1}$. From Tanner [Tan 64, see correction in Tan 78], the ratio, D_g/ε_g, for a variety of soils lies in the range: 0.01 to 0.05 $cm^2 \cdot s^{-1}$. It follows that the diffusion length, ℓ_g, in soil lies in the range: 60 to 154 cm. If we take 0.25 as a typical value for the soil porosity, ε_g, then the corresponding range of values of D_g is 0.0025 to 0.0125 $cm^2 \cdot s^{-1}$. Hence, for $(L/\ell_s) \cong 1$, the flux density ratio given by eq. (1.38) is in the range of 23 to 49. This indicates that one may expect perhaps a 25- to 50-fold diminution in the radon flux density from soils by the introduction of a 10-cm concrete slab (assuming that there is no contribution to the flux density from the concrete itself).

I. E. Conclusions and Recommendations

A basic premise of the model of the transport of radon through a building material such as concrete or phosphogypsum board is that the building material behaves like a porous medium. Any comprehensive study of radon transport in a building material should have two objectives: first, to verify that the transport equations for a porous medium (i.e., Fick's law and Darcy's law) provide a correct description of transport in the material; and if so, second, to measure the transport coefficients and material constants for the material, i.e., measure the diffusion coefficient for radon, the permeability coefficient of air, the porosity of the material, and the production rate of radon per unit pore volume.

In view of the absence of such a comprehensive study for any building material, it is recommended that such studies of a series of concretes and phosphogypsum boards be undertaken.

CHAPTER II

THE ROLE OF THE INTERNAL CHARACTERISTICS OF BUILDING MATERIALS

The transport of radon in building materials can be classified as:

(i) flow, where the fluid (liquid water, air, or water vapor) within the interstitial pore spaces of the material entrains the radon and acts as a carrier; and

(ii) diffusion, where the radon moves relative to the fluid (liquid water, air, or water vapor) in the pores.

Building materials may be subjected to liquid or vapor water or air pressure gradients, osmotic gradients (for example, a dissolved alkali concentration gradient), temperature gradients, and radon gas concentration gradients. These gradients, depending on their magnitude and the corresponding building material porosity, can result in significant flow and diffusion of radon. In addition, since radon is relatively soluble in liquid water[*], mechanisms involving liquid water flow may be significant, depending on the flow rate. The internal characteristics and properties of building materials, such as the location of the radium progenitor of radon in the material and the internal geometry of the pore system, can significantly affect the transport of radon.

This chapter, therefore, reviews and considers: the emanation process from a microstructural viewpoint (Section II.A.); the role of the location and distribution of radium in materials (Section II.B.); and the effects of internal geometry (e.g., pore size, distribution, accessibility and continuity), and the content and state of internal water as they may affect the entrainment and transport of radon in materials (Section II.C.). Because concrete and other cementitious materials are the basic components of most buildings, their characteristics with respect to radon transport are particularly important. The characteristics of hardened concrete are therefore more fully considered and developed as a case study (Section II.D.).

II.A. A Microstructural View of the Emanation Process Within a Material

The importance of the internal characteristics of building materials becomes more apparent when one considers the emanation process from a microstructural viewpoint. Tanner [Tan 64; Tan 78] reviewed and discussed the role and relative importance of internal characteristics as related to radon emanation, entrainment, and transport in the ground. Much of this information is also applicable to building materials. The following microstructural view is almost wholly adopted from the review of Tanner [Tan 78].

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[*] In a closed two-phase air-radon-water system in equilibrium at atmospheric pressure and with the partial pressure of radon approximately 10^{-4} mm Hg, the ratio, concentration of radon dissolved in the liquid water phase divided by concentration of radon in the vapor phase, decreases from 0.5 at 0 °C to approximately 0.1 at 60 °C [McK 74; IUP 79].

The release of radon atoms into the interstitial pore spaces of a material is inherently related to the recoil process following radioactive decay of radium. When radium (^{226}Ra) atoms inside a material decay by α-particle emission, radon (^{222}Rn) is formed as recoiling atomic species. The fate of a recoiling radon atom depends not only on the location of the original radium progenitor, but also on the recoil path and the composition of the material in the path. In air, a recoiling radon atom has a range of approximately 60 µm; and in water, the range is approximately 1000 times shorter. Within minerals of common density, a recoiling radon atom has a range of 20-70 nm.

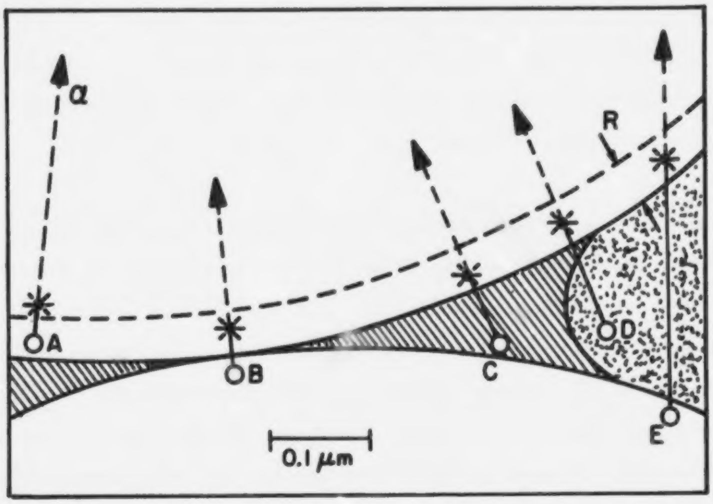

Figure 2.1 Illustration of several scenarios for the release of radon into the interstitial pore spaces of a material. Adopted from Tanner [Tan 78]. For details, see text.

Figure 2.1, adopted from Tanner [Tan 78], illustrates several scenarios for the release of radon into the interstitial pore spaces of a material. The figure represents two spherical grains of 2-µm diameter which are in contact. The interstitial spaces between the grains contain liquid water (lined area) and air (stippled area). The figure shows 5 different paths for recoiling radon atoms directed toward the outer surface of the upper grain. Since the radium progenitor of radon atom A lies at a depth greater than the recoil range R, radon atom A loses all of its recoil energy inside the grain and remains entrapped. Radon atom B recoils out of the upper grain, but embeds into the lower grain. Radon atoms C and D recoil from the upper grain and lose the remainder of their kinetic energy in the pore water and air, respectively. These radon atoms are now free either to diffuse through the pores or to be transported by pore fluid flow. Radon atom E recoils out of the upper grain, loses very little of its recoil energy in the pore air, and buries itself into the lower grain. Radon atoms B and E on penetrating the lower grain, damage the surface and create a "pocket" from which they may escape back into the pores. These mechanisms are discussed in much greater detail by Tanner [Tan 78]. Those radon atoms that terminate their recoil paths in the pores (e.g., atoms C and D) constitute the "direct-recoil fraction" of emanating power; while those that diffuse out of a recoil pocket (e.g., atoms B and E) compose the "indirect-recoil fraction".

Tanner has noted:

> "In compacted natural materials either, 1) the grain sizes are much larger than the recoil range in the grains, so that few recoil atoms escape from the grains in which they originate; or 2) the pores are much smaller than the recoil range in air, so that the recoil paths do not terminate in the pores if the pores are gas filled. The direct-recoil fraction of emanating power is consequently less than 1% in dry compacted granular materials. If the pores contain water or another liquid, however, the range of the recoil atom is only about 0.1 μm [0.06 μm] and the probability that it will stop in the pore is greatly increased. The presence of a liquid in the pores thus increases the direct-recoil fraction of emanating power". [Tan 78]

Hence, the emanating power (particularly, the direct-recoil fraction) of grains within a material and the subsequent transport of radon within the pores of that material is very dependent on the size and structure of the material's pore system and on the composition of the fluids within those pores. These effects will be considered more fully in Section II.C.

II.B. Influence of Radium Content of Materials

Obviously, the amount of radium in a building material is a limiting and critically important intrinsic characteristic of the material with respect to the emanation power of grains in the material, and subsequent radon transport through the material's pore system and exhalation of radon from the material's surface. When considering the microstructural scale of the processes involved, however, it should be apparent that this dependence on the radium concentration is extremely complex. Since, as noted, the recoil range of radon in most common mineral grains is in the range 20-70 nm, the fraction of radon free to move through the pore system of a material will certainly depend on the grain size. But even the relationship between grain size and emanation power is not simple (see, for example, Tanner [Tan 78]); and the effects of the distribution of radium on the surface and within the grains and the distribution of grains in a material are even more confounding. Andrews and Wood [AnW 72], for example, observed two entirely different relationships between emanation power and grain size for two kinds of sandstones because of differing radium distributions in the sandstones.

Known results for the radium content in building materials are summarized in Section III.A. It must be emphasized, however, that existing studies that have attempted to relate the radium content of building materials to radon flux or exhalation rates have taken only a very macroscopic or phenomenological approach. This limitation is perhaps understandable since it would be exceedingly difficult, if not impossible, to attempt to characterize on a microscopic basis the distribution of radium within these materials. The composition of common building materials is highly variable both within a specimen and between specimens, and cannot be considered to be homogeneous on the micron and submicron scale that is of importance.

II.C. Pore Geometry and Water Characteristics

The microstructural viewpoint is similarly useful for understanding the effect of pore geometry and the composition of pore fluids on radon transport in the pore system of a building material. The permeability of building materials (defined as the steady-state flow of a fluid induced by a pressure gradient) is not a simple function of its porosity (ratio of pore volume to total volume), but depends on the size, distribution, accessibility, and continuity of the pores. Accessible pores which run through the specimen primarily control the movement of fluids. Further, the distribution of pore sizes is important. A building material may have a relatively high porosity but a relatively low permeability because of the size and distribution of the pores. For example, in hardened cement gel (discussed in Section II.D.), the gel has a porosity of about 28%, and yet is relatively impermeable because its pores are extremely small. Conversely, a building material, such as natural rock could have a low porosity and a relatively high permeability because some of the pores are accessible, sufficiently large, and continuous through the rock.

If the pore size is on the order of the mean free path of the gas diffusing through the pore, the diffusion will be reduced relative to diffusion in a larger pore [You 70]. Smaller pores tend to retain and fill with liquid water because of the stronger capillary forces associated with smaller pores. Further, the diffusion of gas through the liquid water in the pores is significantly less than diffusion through the air in the pores [Tan 64].

Fluids, including liquid water, water vapor, or air, in the pores may or may not significantly contribute to the transport of radon by flow (fluid acts as a carrier for radon). The contribution of fluid flow mechanisms to the overall radon transport will depend on the relative volume flow rate of the fluid as compared to the transport of radon by diffusion.

As noted previously, the presence of water serves to increase the exhalation of radon from radon producing minerals [Tan 64]. Water in the pores of building materials which contain radium enhances the capture of recoiled radon atoms in the pores (after the atoms escape from the pore wall and enter the pore, see fig. 2.1). When a recoiling radon atom escapes from a solid pore wall and encounters liquid water, it will be slowed down considerably as compared to the case when no water is present. The extent to which the radon atom is slowed down or stopped depends on the size of the pore and the amount of water in the pore. Based on the mechanisms discussed by Tanner (fig. 2.1), the higher the fraction of liquid water in the pores and the larger the fraction of water-filled (partially or totally filled) pores in the material, the greater the probability of radon being entrained in the pores and free to move.

In summary, one may conclude that pore accessibility, continuity, size distribution and water content are major factors that affect fluid transport, and thus radon transport, through building materials. Liquid water in the pores will significantly decrease the diffusion of radon as compared to the diffusion of radon in air-filled pores. Pore water content and size distribution appear to be major parameters in affecting radon entrainment in the pores. To illustrate the effects of the variables being discussed, a building material may have a high radium content and a high porosity, but may have low exhalation of radon due to pore size distribution (many small pores) and have low radon capture due to a lack of water in the pores.

II.D. A Case Study: Hardened Concrete

This section, using hardened concrete as an important and representative example of a building material, more fully reviews the role of internal characteristics in affecting radon entrainment and transport. It includes definitions of the important properties of hardened concrete, typical values for these properties, and a discussion of their role.

II.D.1. Characteristics of Hardened Concrete

a) Composition

Hardened concrete consists primarily of mineral rock (aggregate) and hardened cement paste which is used to coat and bond the aggregate together. By volume, about 60 to 80% of concrete is aggregate

[PCA 79]. In addition to the aggregate and paste, there is considerable void space. The void spaces (pores) contain no solid material but are filled with fluids, typically air, water vapor, liquid water (which contains dissolved alkalies), or a combination of one or more of these.

b) Porosity of Hardened Cement Paste

The void spaces in the hardened cement paste and concrete can be classified [Woo 68] as shown in Table 2.1. Capillaries and gel pores deserve special discussion. Capillaries are cavities which were originally water-filled spaces in unhardened concrete. As hydration proceeds some of the water is used to form hydrated cement gel. The remaining space not occupied by cement gel is termed the capillary porosity. Figure 2.2 taken from Powers [Pow 62], illustrates a simplified model of the cement gel structure and void space.

Table 2.1

Void Classification and Characteristics for Hardened Concrete [Woo 68].

Class	Size	Shape	Volume Percent
Entrapped air	Barely Visible to about 1/2 in (1 cm) or more	Irregular	Minimum of 1% by volume of concrete
Entrained air	About 0.003 to 0.004 in (0.008 to 0.01 cm)	Nearly Spherical	Up to 5% or more by volume of concrete
Capillaries	From about 10^2 to 10^4 Å [VeH 69] (1 Å = 10^{-8} cm)	Irregular with tendency to interconnect	Typically 15-40% or more by volume of the cement paste [Pow 78]
Gel Pores	On the order of 18 Å		28% by volume of hydrated cement gel

The hydration and hardening of cement has been studied at length (see, for example, Powers [Pow 62] and Williamson [Wil 73]). Hardened cement paste consists of the products of hydration (gel and calcium hydroxide crystals), unhydrated cement, and the capillaries. Hydrated cement gel is the "solid" part of hardened cement paste, including calcium hydroxide crystals, but excluding capillaries and unhydrated cement. As hydration proceeds, part of the water-filled space becomes filled with solid cement gel, constricting the capillaries. Whether complete isolation of the capillaries can occur, even with low water/cement ratios and complete hydration, is not known. Recent evidence [VeH 69, CoV 76] suggests that, although the capillaries become constricted, they never become isolated.

The pores of the cement gel are very small and numerous and usually contain water. The gel pores have a characteristic porosity of about 28% by volume of the cement gel. The capillary porosity of hardened cement paste (excluding gel porosity) in ordinary concrete ranges from about 15 to 40% or more by volume of the cement paste [Pow 78]. Thus the total porosity (gel and capillary) typically ranges from about 40 to 55% or more by volume of the cement paste.

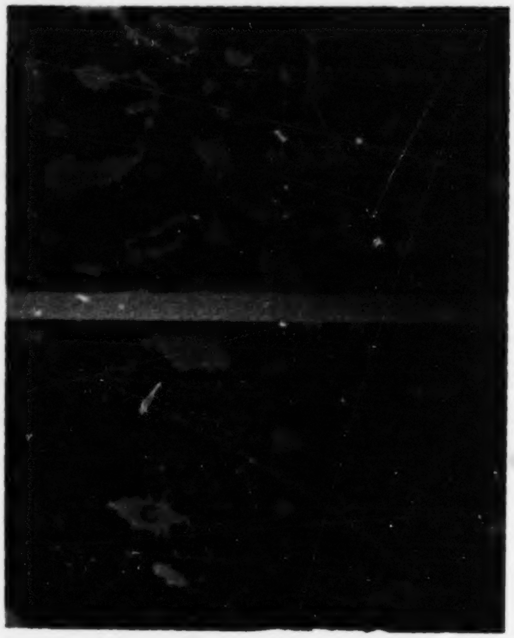

Figure 2.2. Illustration of a simplified model of paste structure in cement gel. Masses of randomly oriented groups of black lines represent cement gel. Spaces like those marked C represent capillary cavities. Upper drawing represents mature paste, water/cement ratio = 0.5, capillary porosity 20%; lower drawing represents nearly mature paste, water/cement ratio = 0.3, capillary porosity 7%. From Powers [Pow 62].

c) Porosity of Aggregates and Concrete

The porosity of most acceptable natural aggregate lies below 10% and commonly below 3% [Woo 68], in contrast to the 15 to 40% or more (capillary porosity) for hardened cement paste. Although the porosity of the aggregate can be considerably less than that of the hardened cement paste, the water permeability of the aggregate can be on the order of, or even higher than that found for mature cement paste with water/cement ratios in the range of about 0.4 to 0.7. This is because on the average the pore size in aggregates is considerably larger (though larger pores in aggregates are usually present in much smaller proportion) than that for hardened cement paste [Woo 68]. Thus, the effective porosity of concrete is a weighted average of the porosity of the aggregate and hardened cement paste.

d) Measurement of Porosity

Copeland and Verbeck [CoV 76] and Verbeck and Helmuth [VeH 69] reviewed the determination of porosity and pore-size distribution. Pore-size distributions can be calculated, provided an assumption concerning pore shape or pore "hydraulic radius" (pore volume/pore wall area) is made. Two methods are typically used:

i) porosimetry, in which the volume of mercury that is forced into the porous sytem is measured as a function of the applied pressure; and

ii) capillary condensation of vapors from either an adsorption or desorption isotherm.

Sorption results are limited to pore diameters less than about 300 Å (0.03 μm) while the mercury porosimetry method can measure pore diameters as high as 1 μm [CoV 76]. Copeland and Verbeck [CoV 76] indicate that the significance of pore-size distribution as determined by either technique needs additional clarification.

e) Water Retention in Capillaries

Woods [Woo 68] points out that the tenacity with which the various classes of voids retain water is inversely proportional to their size. Water in the gel pores is so firmly fixed that it does not evaporate under drying conditions that will remove nearly all the capillary water; yet this water, to a limited extent, participates in the flow of water [Pow 62]. At the other extreme, are the larger entrapped and entrained air voids which have little propensity for retaining water and thus are usually empty. Capillaries, which represent the intermediate void pore size, have an intermediate affinity for water. Thus, a competition exists between the various void sizes for the available water. It has been observed that the larger pores (including entrained air voids) within a body of concrete subjected to wetting, will remain unfilled, even in water-soaked concrete [Woo 68].

f) The State of Water in Hardened Cement Paste

Water in hardened cement paste can be arbitarily classified as "evaporable" and "non-evaporable" (that is, water not removed under a standard drying condition, which is commonly performed in a vacuum over dry ice (-79 °C) [VeH 69]). The drying is intended to remove nearly all the "physically adsorbed" water and not the "chemically bound" water of hydration. The "evaporable" water can be further classified as "gel" water and "capillary" water, where "capillary" water is defined as the difference between the total evaporated water of a paste and its gel water content. Copeland and Verbeck [CoV 76] classify the water in hardened cement paste in more detail as:

i) water chemically combined in the hydration products;
ii) hydrate water in crystallites;
iii) adsorbed water on solid surfaces; and
iv) water "in capillary pores large enough to have space outside the force fields of solid surfaces."

They indicate that experimental differentiation among these classes of water has not been achieved except for the case of the chemically combined water which can be approximately distinguished from the other three classes. They also indicate, however, that some hydrate water is removed under drying and "part of the molecular water may remain". For example, using quasi-elastic neutron scattering, Harris et al. [HWL 74] observed that saturated cement pastes had free water fractions smaller than the values determined by simple drying weight-loss experiments. They conclude from their data that free and combined water are lost simultaneously during evacuation (drying).

Copeland and Verbeck [CoV 76] discuss the state of information on the density and thickness of the multilayers of adsorbed water on the capillary walls caused by the interaction between the solid phase and water at the interface. Exact values for the specific volume of the adsorbed water are not known, but estimates indicate that in saturated pastes they are about the same as, or a little larger than, the values of pure bulk liquid water. Copeland and Verbeck [CoV 76] indicate that the significance of knowing the specific volume of the adsorbed water is that the pore volume of saturated hardened paste can be determined with relatively little error by determining the volume of evaporable water in a unit volume of saturated paste. There is reasonable evidence suggesting that more than one layer of adsorbed water exists in capillary pores of concrete, provided some degree of saturation exists in the pores [Pow 62].

II.D.2. Movement of Pore Fluids in Concrete

Concrete can be subjected to various fluid (liquid water, water vapor or air) pressure gradients. These gradients can result in the flow and diffusion of radon in the liquid water, water vapor, or air of the concrete.

a) Movement of Water

Although the cement gel has a porosity of about 28%, its permeability is of the order of 10^{-13} cm·s^{-1} [PCH 54]. The cement paste as a whole, including capillaries, in mature, ordinary concrete can be considerably more permeable (several orders of magnitude or more) than the gel [PCH 54], since water can flow through the capillaries more easily than through the extremely small gel pores. Thus the permeability of hardened cement paste is controlled by its capillary porosity. Verbeck and Helmuth [VeH 69] present for hardened cement paste a relationship between permeability and total porosity (gel and capillary porosity), water viscosity, and temperature.

As discussed earlier, the permeability of the aggregates may equal or exceed that of the cement paste, even though the porosity of the aggregate may be considerably lower than that of the paste. Thus the permeability of concrete will be dependent on the permeability of the hardened cement paste, the aggregate, and the aggregate-cement paste interface. If the permeability of the aggregate is lower than that of the cement paste, the presence of aggregate (60 to 80% by volume of concrete) will reduce the area through which flow can occur. In effect, the length of the flow path has been increased, since the flow has to circumvent the aggregate.

The most likely mechanism to cause water migration through a concrete wall or floor slab is that resulting from a relative humidity gradient across the thickness. The primary mechanism is believed to be that as described by the following Building Research Advisory Board statement [NAS 58] on transfer of moisture through concrete slabs-on-ground:

> "The Committee believes that moisture is transferred through partially dry concrete in the adsorbed or condensed state by surface diffusion and does not move as vapor through the concrete. Also, where good practice has been followed in the design and curing of concrete, it is believed that moisture does not move through the concrete by capillarity as it is usually understood."

A relevant Portland Cement Association laboratory study [Bre 65], showed that measurable moisture movement occurred through 4-inch (10.2 cm) thick concrete slabs exposed to liquid water on one side and 50% relative humidity on the other. Both sides of the slabs were maintained at 73 °F (23 °C). Volume flow rates (volume of liquid water) measured over a wide range of water/cement ratio values were on the order of 10^{-8} cm^{3}·s^{-1}·cm^{-2} through the 4-inch (10.2 cm) thickness. These values correspond to approximate steady state flow conditions attained after a year of exposure.

b) Movement of Air

As with the permeability of liquid water in concrete in which the pores are filled with liquid water, the flow rate of air depends on the section thickness and the pressure gradient [Nev 71]. There is, however, no unique relationship between the permeability for air and water for all concretes. The steady state flow rate of air can be achieved in several hours depending on the porosity and pore size distribution. Neville [Nev 71] lists some data on the flow rate of air for concrete with a water/cement ratio of 0.62. The rate of flow values, in units of $in^3 \cdot (lb \cdot in^{-2})^{-1} \cdot h^{-1} \cdot ft^{-2}$ ranged from 0.58 to 0.24 [i.e, 0.015 to 0.0062 $cm^3 \cdot (Pa)^{-1} \cdot h^{-1} \cdot m^{-2}$] for the thickness ranging from 4 to 9 inches (10 to 23 cm), respectively. Loughborough [Lou 66] reported air permeability coefficient values for concrete at various moisture contents and having a water/cement ratio of 0.50. The values ranged from 0.04 to 0.23 expressed as the volume of air in cubic inches (1 in^3=16.4 cm^3), at atmospheric pressure and zero °C, that in 1 minute flows through a 1 square foot (0.0929 m^2) area of 1 inch (2.54 cm) thick concrete, at a pressure differential of 1 psi (6895. Pa) across the specimen. Four inch (10.2 cm) thick specimens subjected to a 14 psi (96530. Pa) pressure difference were used. With a pressure difference of 25 mm Hg and a concrete thickness of 4 inches (10.2 cm), the corresponding volume flow rate based on Neville's and Loughborough's data is of the order of 10^{-6} $cm^3 \cdot s^{-1} \cdot cm^{-2}$.

II.D.3. Effect of Concrete Properties on Radon Transport

a) Transport by Flow

Based on the data presented, air can act as a carrier for radon gas. The volume flow rate of air corresponding to a 25 mm Hg air pressure difference across a 4-inch (10.2 cm) concrete thickness is of the order of 10^{-6} $cm^3 \cdot s^{-1} \cdot cm^{-2}$. Liquid adsorbed water is believed to act as a carrier where there is a relative humidity gradient across the concrete thickness. With a 4-inch (10.2 cm) concrete thickness at 73 °F (23 °C), the volume flow rate (volume of liquid water) corresponding to one face wet and the other face at 50% relative humidity, is of the order of 10^{-8} $cm^3 \cdot s^{-1} \cdot cm^{-2}$, which is several orders of magnitude smaller than the air flow rate. The relative significance of radon transport by flow resulting from air or moisture movement would depend on the intensity and proportion of time the driving force (air pressure or relative humidity gradient) acts across the concrete thickness. In field conditions an air pressure difference of 25 mm Hg will not always be present. In contrast, a relative humidity gradient can often be present much of the time. Thus, further research is needed to determine which flow mechanism (air or moisture) would contribute more to the transport of radon by flow.

b) Transport by Diffusion

Considering only diffusion, the presence of liquid water in the pores significantly reduces radon diffusion, due to the large difference between diffusion coefficients for radon in air (on the order of 10^{-1} $cm^2 \cdot s^{-1}$) and liquid water (on the order of 10^{-5} $cm^2 \cdot s^{-1}$) [Tan 64]. Thus, radon diffusion in pores partially filled with liquid water would probably be primarily controlled by the diffusion of radon in the air rather than the diffusion in the adsorbed water layers.

II.E. Conclusions

The effects of the internal characteristics of building materials (including radium content and distribution, internal geometry, pore size and distribution, and water content and state) on the entrainment and transport of radon through the pore system have been discussed. As indicated, these effects are exceedingly complex. Radon transport by flow, resulting from the movement of air and moisture through the pore system, is possible. The relative contribution of the air and moisture movement to radon transport would depend on the intensity and proportion of time the driving force (air pressure or relative humidity gradient) acts across the material thickness. The presence of liquid water in the pores can significantly reduce the diffusion of radon, but at the same time, the presence of liquid water in the pores can enhance the entrainment of recoiling radon atoms.

CHAPTER III

A REVIEW OF EXPERIMENTAL DATA

This chapter considers the radon exhalation process from a more macroscopic, phenomenological viewpoint. In part, it focuses on the question: Is it possible to predict or estimate radon exhalation from building materials based on existing data? Three categories of data have been reviewed and summarized:

A. radium concentrations in concrete and other building materials;

B. radon exhalation from soils, concretes and other building materials; and

C. radon exhalation as a function of meteorological parameters.

Data were selected to show typical values or ranges of values. In addition, values for radium content and meteorological parameters are addressed in terms of their potential effect on radon flux densities and exhalation rates. Since it is often of interest in a review such as this, the criterion used to select the data is indicated in each case. Lastly, in Section III.D., these data are summarized in tabular form to highlight not only the expected range of values for radon flux densities and exhalation rates, but also to identify some of the most critical factors.

Although this chapter is based on a survey of several hundred papers, a number of extensive review articles have also been relied on. These reviews, such as those by UNSCEAR [UN 77; UN 80], CRCPD [CRC 78], Harley [Har 78], Eadie [Ead 75], Wilkening, et al. [WCS 72], and Tanner [Tan 64; Tan 78], cover virtually all of the literature relevant to this chapter. The survey made for this study and the previous reviews generally concur that there is very little information that is directly usable for predicting radon flux densities or exhalation rates.

Before discussing the existing data, however, it may be useful to evaluate the magnitude of the radon flux densities which may be considered to be a significant contributor to the indoor radon concentration.* Consider a single-room structure 2.5 m in height and with a 10-cm concrete slab floor located directly on a soil surface. A typical radon density flux from dry soil is 0.43 $\mathrm{pCi \cdot m^{-2} \cdot s^{-1}}$ (i.e., 0.75 $\mathrm{atoms \cdot cm^{-2} \cdot s^{-1}}$) [WCS 72]. If all of the exhaled radon from the soil enters the room without diminution by the floor, then approximately 1500 pCi of radon will enter through each m^2 per hour. If this is the only source of indoor radon, then, with a ventilation rate of $\lambda_v = 0.5\ h^{-1}$, the steady-state concentration of radon in the room, given by the first term of eq. (1.2), would be 1.2 $pCi \cdot L^{-1}$. Assuming an equilibrium factor of 0.5, this radon concentration corresponds to an indoor working level of 0.006 WL*. This is somewhat lower than levels currently conceived to be significant and possibly warranting remedial action (e.g., 0.01-0.02 WL). In addition, the reduction of the radon flux density from the soil due to an intact 10-cm concrete slab is roughly a factor of 25 to 50 (see Section I.D.8). Hence, it appears that the radon flux density from soils near the worldwide average of 0.43 $pCi \cdot m^{-2} \cdot s^{-1}$ should not be of great concern as a source of indoor radon. This may not be applicable in buildings that have cracks or unsealed service entrances in the foundation, or have considerably reduced ventilation rates. Also, some "technologically-enhanced" soils and radium-bearing building materials exhibit considerably elevated radon flux densities and may be significant sources to indoor radon concentrations. There are, of course, other contributing sources (see introduction to Chapter I) which may exacerbate the significance of radon exhaled from soils and building materials. Radon flux densities from soils and building materials are commonly within a factor of 10 of 5 $pCi \cdot m^{-2} \cdot s^{-1}$ which may lead to indoor levels within a factor of 10 of 0.02 WL. This suggests that field measurements should be accurate to at least a factor of 2. Furthermore, on this same basis, the influence of a given variable affecting the flux density might be considered significant if a typical change in that variable results in a 10-20% change in the flux density. In this way, even if the individual effects of the possible affecting variables add together, the overall effect on the radon flux density could still be relatively unimportant. Reported variations of more than 10-20% (e.g., due to changing atmospheric conditions) are common. Descriptions of how these observations can be used to estimate or predict radon flux densities are much less common. Generally, only a qualitative understanding of the dependence of the radon flux density on a given variable can be inferred because in most studies other variables were not controlled.

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*This evaluation was made under the assumption that the radon flux density from open soil provides an upper limit on the radon from soil that can enter a building. W. W. Nazaroff (Lawrence Berkeley Laboratory, Berkeley, California) has suggested that this assumption may not be valid, and has offered an unpublished alternative "stack effect" mechanism that leads to different conclusions. Readers may wish to contact him directly for further details.

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*The WL unit is defined as any combination of the short-lived radon progeny (^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po) in one liter of air that will result in the ultimate emission of 1.3×10^5 MeV of alpha-particle energy [UN 77]. The numerical value of the WL (1.3×10^5 MeV per liter) is derived from the total α-particle energy ultimately emitted in the decay of the short-lived ^{222}Rn progeny that are in radioactive equilibrium with 100 pCi of ^{222}Rn per liter of air. Although this numerical value is derived with the assumption of radioactive equilibrium (i.e., 100 $pCi \cdot L^{-1}$ of each ^{222}Rn progeny), the unit is applicable for any mixture of short-lived ^{222}Rn progeny including nonequilibrium situations. Hence, with an equilibrium factor of 0.5, the working level corresponding to a radon concentration of 1 $pCi \cdot L^{-1}$ is 0.005 WL.

III.A. Radium Content of Building Materials

Values for the radium content of building materials have been reported or summarized by Hultqvist [Hul 56], Krisiuk, *et al*. [KTS 71], Eadie [Ead 75], Hamilton [Ham 71], Kolb [Kol 74], CRCPD [CRC 78], UNSCEAR [UN 77], Harley [Har 78], Fitzgerald and Sensintaffer [FiS 78], and others. Despite this seemingly substantial list, the amount of available information is actually relatively scant. Eadie [Ead 75], for example, in reviewing 275 references pertaining to radioactivity in building materials states as his first "conclusion" that "the article by Hamilton [Ham 71] is the only significant report of data on the radioactivity of specific building materials." Similarly, Harley [Har 78] states: "In reviewing the papers available, it is apparent that relatively few present data for amounts of radioactive substances and for the radiation exposure within the building. More data of this type would be useful......" A Task Force of the CRCPD [CRC 78] noted, however, that by 1978 there had been analyses of the radioactivity concentrations of commonly used building materials in seven countries, but not in the United States. The most recent summaries of results for the radium content of building materials appear in the UNSCEAR compilations [UN 77; UN 80].

Table 3.1, adapted from Harley [Har 78], summarizes the results for some select building materials. The data were selected to better illustrate typical ranges and the distribution for a few important materials, as well as to facilitate comparisons between different types of material (bricks, concretes, etc.). Thus, for example, the "clay brick" in the U.K., the "red brick" in the U.S.S.R., and the "brick" in West Germany had radium concentrations of 1.4, 1.5, and 1.7 $\text{pCi}\cdot\text{g}^{-1}$, respectively; while the "silica brick" in the U.S.S.R. and "silicate brick" in the U.K. were about a factor of 3 to 7 less (0.5 and 0.2 $\text{pCi}\cdot\text{g}^{-1}$, respectively). Similar comparisons can be made for other building materials such as

Table 3.1

Radium Concentrations in Selected Building Materials

Reference	Material	^{226}Ra Concentration ($\text{pCi}\cdot\text{g}^{-1}$)
Hamilton [Ham 71] (U.K.)	Clay brick	1.4
	Silicate brick (Gravel)	0.2
	Granite	2.4
	Aerated concrete	2.4
	Natural gypsum	0.6
	Concrete block (Fly Ash)	(0.2-2)
Krisiuk, *et al*. [KTS 71] (U.S.S.R.)	Red brick	1.5
	Silica brick	0.5
	Light concrete	2.0
	Granite	3.0
	Sand	0.4-1
	Cement	0.7
Kolb [Kol 74] (W. Germany)	Granite	2.8
	Brick	1.7
	Sand, gravel	0.4
	Cement	0.5
	Natural Gypsum	0.5
	Concrete	0.6

concretes which range from 0.6 to 2.4 $pCi \cdot g^{-1}$. As a result of these variations, typical values for the radon flux density from these materials vary from roughly 30% to factors of 2 or more. In addition, many components of building materials (e.g., the aggregate in concrete) are locally supplied, and, as noted by the CRCPD Task Force, the "data on radionuclide concentrations in many construction materials showed large variability from region to region" [CRC 78].

The radioactivity content of "phosphogypsum," a potentially important building material, has been reviewed by Fitzgerald and Sensintaffer [FiS 78]. Massive quantities of this by-product of phosphate processing operations are available, and pose a major disposal problem. Its use in building materials has not been widespread in the U.S., but has been contemplated. The radium concentration of phosphogypsum, as shown in Table 3.2, is approximately an order of magnitude greater than that for naturally-mined gypsum. The National Radiological Protection Board in the United Kingdom recommended that sources of raw materials having concentrations "significantly in excess of 25 $pCi \cdot g^{-1}$ should be avoided" [ODS 72]. The variability in radium content and radon exhalation rates from phosphogypsum products should be studied more closely since, as seen in Table 3.2, average radium concentrations are at or near this limit.

Table 3.2

Radium Concentrations in Phosphogypsum [FiS 78]

Phosphogypsum Source	Average ^{226}Ra Concentration ($pCi \cdot g^{-1}$)
Florida	33
Morocco	25
Idaho	23
Khoaribga	17

III.B. Radon Exhalation from Soils and Building Materials

Radon exhalation from soil has been studied extensively since the early part of this century; the first value being reported in 1909. Numerous results are available, and Wilkening, et al. [WCS 72] reviewed the data from the standpoint of obtaining a worldwide average radon flux density. Based on a review of nearly 1000 measurements of the radon flux density from soils of various types at locations across the earth, they obtained a worldwide overall average of 0.43 $pCi \cdot m^{-2} \cdot s^{-1}$ with a range from 0.006 to 1.4 $pCi \cdot m^{-2} \cdot s^{-1}$ [WCS 72]. Table 3.3, abstracted from this paper, tabulates some of the results for the radon flux density from soils in the U.S. Typical values vary by a factor of 2-3, and are not substantially different from the worldwide average. For example, the overall mean for the sampled New Mexico locations is 0.65 ± 0.19 $pCi \cdot m^{-2} \cdot s^{-1}$ compared with 0.28 ± 0.09 $pCi \cdot m^{-2} \cdot s^{-1}$ for Texas, 0.19 ± 0.06 $pCi \cdot m^{-2} \cdot s^{-1}$ for Alaska and 0.30 ± 0.09 $pCi \cdot m^{-2} \cdot s^{-1}$ for Hawaii.

Table 3.3

Radon Flux Density from Soils in the U.S.

Location[a]	Soil Type	Measurement Method[c]	^{222}Rn Flux Density ($pCi \cdot m^{-2} \cdot s^{-1}$)
Alaska			
Tanana basin (14)	Podsolic	A	0.13 ± 0.04
Tanana basin (3)	Podsolic	VP	0.21 ± 0.06
Yukon basin (1)	Podsolic	VP	0.22 ± 0.07
Hawaii			
Deep soils (6)	Latosolic	A	0.85 ± 0.23
Thin soils (18)	Mountain	A	0.05 ± 0.01
Lavas (5)	Mountain	A	0.06 ± 0.002
Illinois			
Champaign County (472)	Chernozemic	F	1.4
Argonne (8)	Chernozemic	F	0.57 ± 0.05
Massachusetts			
Lincoln (10)	Podsolic	A	1.4
Nevada			
Yucca Flats	Desertic	A	0.48
New Mexico			
Socorro (10)	Desertic	F	0.91 ± 0.09
Socorro (6)	Desertic	F	1.0 ± 0.3
Valley (130)[b]	Desertic	A	0.33 ± 0.23
Plains (4)	Desertic	A	1.2 ± 0.3
Plains (8)[b]	Desertic	A	0.54 ± 0.16
Mountains (10)[b]	Mountain	A	0.18 ± 0.06
Others (3)	Desertic	A	0.62 ± 0.17
Socorro (3)	Desertic	VP	0.56 ± 0.16
Texas			
Plains (3)	Chernozemic	A	0.41 ± 0.12
Central (3)	Chernozemic	A	0.20 ± 0.06
Coastal (3)	Latosolic	A	0.23 ± 0.07

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[a] Numbers in parentheses give number of measurements.
[b] Two sites.
[c] A-accumulator; VP-vertical profile; F-flow (see Section IV.C.1.).

Wilkening, et al. [WCS 72] noted that "wide variations occur in radon flux density measured at different sites, at different times at a given site, and by different methods employed simultaneously at the same site." They listed 6 factors that influence the flux density at a given location:

(i) the radium concentration in the soil and rocks (see Sections II.B. and III.A.);

(ii) the emanating power of mineral grains or soil particles - i.e., the ability of the radon atoms formed by the decay of radium to escape from the grains (see Section II.A.);

(iii) the porosity and equivalent depth (diffusion length) of the soil which determines the transport characteristics of radon in the interstitial pore spaces of the soil (see Chapter I);

(iv) the state of vegetative cover;

(v) water and or ice involvement (see Sections II.C. and III.C.1.); and

(vi) variations in atmospheric characteristics that influence transport processes between the soil gas and lower layers of the atmosphere (see Chapter I and Section III.C.).

Radon exhalation from "technologically-enhanced" materials is much more variable. Bernhardt, et al. [BJK 75], for example, reported values for radon flux density from uranium mill tailings at four widely separated areas. The values range, as shown in Table 3.4, over three orders of magnitude.

Table 3.4

Radon Flux Density from Uranium Mill Tailings [BJK 75]

Location of Uranium Mill Tailings[a]	Range of Values for Radon Flux Density ($pCi \cdot m^{-2} \cdot s^{-1}$)
Shiprock, NM (8)	440 - 2200
Salt Lake City, UT (20)	110 - 780
Lakeview, OR (10)	0.76 - 31
Boulder City, NV (4)	0.77 - 11

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[a] Numbers in parentheses give number of samples.

Horton [Hor 79] similarly found the radon flux density from inactive uranium mill tailings piles and from phosphogypsum piles to vary over several orders of magnitude (Table 3.5).

Table 3.5

Typical Radon Flux Density from Inactive Uranium Mill Tailings and Phosphogypsum Piles [Hor 79]

Source	Range of Values for Radon Flux Density ($pCi \cdot m^{-2} \cdot s^{-1}$)
Inactive uranium mill tailings piles	60 - 1200
Phosphogypsum piles	2 - 120

Few data for radon flux densities or exhalation rates from building materials are available. The 1977 UNSCEAR report [UN 77], for example, cites only four references. Table 3.6, taken from UNSCEAR [UN 77], summarizes the values for various materials. Because of the large variability in radium content even within a given type of material, the most useful data are those which relate the radon flux density or exhalation rate to the radium concentration of the material (as in Table 3.6). This type of information is not always available, however, and the utility of the data is thereby more limited.

Table 3.6

Radon Flux Density Normalized to Radium Concentration for Various Materials [UN 77]

Material	Comments	^{222}Rn Flux Density per Unit Concentration of ^{226}Ra ($pCi\cdot m^{-2}\cdot s^{-1}$ per $pCi\cdot g^{-1}$)
Phosphogypsum	internal walls, 7.6-cm thick	0.01
Phosphogypsum	ceilings, 1.3-cm thick	0.001
Uranium mill tailings	10-cm thick	0.2
Uranium mill tailings	"infinite" thickness	1.6
Soil	"infinite" thickness	0.5
Concrete	10-cm thick	0.005
Light Concrete	20-cm thick	0.02
Heavy Concrete	8-cm thick	0.01

Two more recent studies of radon exhalation from building materials have been reported by Jonassen and McLaughlin [JoM 76; JoM 78] and by Pensko and Stpiczynska [PeS 78]. In the first study, Jonassen and McLaughlin measured the exhalation rates from 14 different types of materials, and reported the values on surface area, volume, and mass bases. They noted that in some cases the material should be characterized in terms of the exhalation rate per unit surface area, (i.e., flux density) while in other cases it appears to be more relevant to describe the material in terms of the exhalation rate per unit mass or exhalation rate per unit volume. They also indicated that it is not possible to predict *a priori* "which if any of these exhalation units is appropriate to use in scaling up the results obtained from samples to a room with walls made of the same material [JoM 76]." Table 3.7 tabulates the radon flux density for the 14 materials studied in their work. For illustrative purposes, it may be useful to compare the flux density from some of the concretes to that expected from soil. For soil having a flux density of 0.4 $pCi\cdot m^{-2}\cdot s^{-1}$ below a 10-cm concrete slab floor, the contribution to the flux density at the surface of the floor could be 0.01 $pCi\cdot m^{-2}\cdot s^{-1}$ (assuming a 40-fold diminution by the floor). Thus, the flux density from any of the first six concretes listed in Table 3.7 is within a factor of 5 and nearly comparable to the flux density through the slab from the underlying soil.

Table 3.7

Radon Flux Density from Various Building Materials [JoM 78]

Material	^{222}Rn Flux Density ($pCi \cdot m^{-2} \cdot s^{-1}$)
1. Ordinary concrete, gravel and sand from the sea, Danish deposits	0.0095
2. Ordinary concrete, gravel and sand from pits, Danish deposits	0.0076
3. Ordinary concrete, unknown composition	0.0084
4. Ordinary concrete, unknown composition	0.0104
5. Light weight concrete, Swedish origin alum shale, old type	0.232
6. Light weight concrete, Swedish origin alum shale, new type	0.082
7. Light weight concrete, Danish origin, clay based	0.0018
8. Light weight concrete, Danish origin, clay based	0.0012
9. Expanded clay concrete, LECA	0.0012
10. Bricks, solid type	0.0013
11. Bricks, cavity type	0.000055
12. Chipboard	0.000019
13. Fiberboard	0.000030
14. Gypsum board	0.000035

In the second study, Pensko and Stpiczynska [PeS 78] evaluated several types of by-products that may be utilized in building materials as well as some conventional building materials. Their results for both the radium concentrations and emanation powers are provided in Table 3.8.

Table 3.8

Emanation Power of Various Building Materials [PeS 78]

Material	Number of Samples	^{226}Ra Concentration ($pCi \cdot g^{-1}$)		Emanation Power (%)	
		Range	Mean	Range	Mean
Fly-ash	33	1.7-3.9	2.6	0.20-1.16	0.54
Slag	11	1.0-3.6	1.8	0.24-1.53	0.70
Phosphogypsum	3	0.7-19	11.8	3.5-19.7	12.0
Red brick	1		0.54	5.1	
Silica brick	1		0.18		16.3
Portland Cement	1		0.65		4.1
Soil	5	0.11-0.51	0.27	12.1-50.1	28.9

Very recently, Nero et al. [NBH 80] at Lawrence Berkeley Laboratory, reported results for the exhalation rate per unit mass from about 100 ordinary concrete samples which originated from 10 different locations throughout the U.S. Values ranged between 0.2 and 2.0 $pCi \cdot kg^{-1} \cdot h^{-1}$.

In addition to these studies, Culot et al. [COS 76] at Grand Junction and Dilworth, et al. [DSM 78] at Elliot Lake measured the radon flux density from concrete slabs overlying "technologically-enhanced" materials. Culot, et al. obtained a flux density ranging from 0.21 to 0.26 $pCi \cdot m^{-2} \cdot s^{-1}$ through a 20-cm thick slab adjacent to uranium mill tailings having a radon concentration of 40,000 $pCi \cdot L^{-1}$. In the work at Elliot Lake, the radon concentration in the underlying soil was approximately two orders of magnitude less than that at Grand Junction, but the radon flux density from the concrete slab was only a factor of two less. In the Elliot Lake Study, it was assumed that a substantial fraction of the flux density was due to contributions from the concrete itself.

III.C. Meteorological Factors Affecting Radon Exhalation

The influence of meteorological factors on the exhalation of radon is an important consideration that should not be overlooked in any predictive model. Although nearly 60 papers which address the effects of external factors on radon exhalation have been surveyed, very few contain sufficient quantitative data that are directly usable. This review focuses exclusively on those few studies which do provide quantitative results.

In one of the most comprehensive studies, Steinhausler [Ste 75] evaluated the dependence of 24 meteorological variables on the indoor radon concentrations in private and public buildings. He found large daily and seasonal fluctuations even under constant ventilation conditions. Using a multiple-regression analysis technique, he was able to identify those variables most strongly correlated to changes in the indoor concentrations. These were: the barometric pressure; the soil temperature; daily mean atmospheric temperature; daily mean wind speed; the relative humidity; the daily atmospheric temperature range; and vertical temperature gradient.

Although meteorological effects may be important, it must be emphasized that the internal characteristics of materials (e.g., radium concentration, porosity, water content, etc.) are usually more significant. Styro [Sty 68], for example, concluded that the influence of meteorological variables on radon exhalation from soils "has only a minor role and that the radon flux (density) is predominantly determined by the characteristics of the soil" (see also, Wilkening, et al. [WCS 72]). Auxier [Aux 73] noted that this was also the case for concrete materials, and concluded that the most important parameter influencing radon exhalation from building materials (for a given radium concentration) is the moisture content of the material (see also Kovach [Kov 46], and Wilkening and Hand [WiH 60]).

III.C.1. Moisture Content of Materials and Humidity Effects

Although moisture effects, particularly the influence of precipitation on radon exhalation from soils, have been discussed in a number of studies [Oka 56; JaH 58; EKS 62; Pea 65; PeJ 65; Pea 67; IsH 70; SNS 70; MeM 73], quantitative results obtained with other factors held constant are scant.

Cox, et al. [CBK 70] examined the radon concentrations in ground-level air at Cincinnati over a 7-year period, and derived two alternative 2-parameter equations for predicting outdoor radon concentrations. The two parameters used to obtain the average monthly morning radon concentration (C), in units of $pCi \cdot m^{-3}$, were the atmospheric thermal stability (S), in °F/27 m, and the total moisture detention in soil (TMD), in units of mm*. The two equations obtained were:

$$C = 139\ (S) + 93{,}000(TMD)^{-1} - 14 \qquad (3.1)$$

and

$$\ln C = 0.332\ (S) - 0.00384(TMD) + 6.877 \qquad (3.2)$$

both of which had a squared correlation coefficient of $r^2=0.89$. As shown in figure 3.1, the radon concentrations, which varied over a factor of about 6, were highly correlated with the TMD. In fact, the radon concentration C could be predicted almost as well using only the TMD in a 1-parameter equation,

$$C = -44 + 114{,}000\ (TMD)^{-1}\ , \qquad (3.3)$$

shown as the solid curve in figure 3.1. Equation (3.3) which has a squared correlation coefficient of $r^2=0.84$ has an overall predictive power nearly equal to eqs. (3.1) and (3.2). The relative significance of the moisture content of soil can be readily seen by differentiating eq. (3.3). At a typical TMD value of 200 mm,

$$\Delta C = -2.85\ \Delta(TMD)$$

which indicates that a 20% change in TMD gives rise to nearly a 60% change in C. At a TMD value of 100 mm, the effect is even greater; a 20% change in TMD results in a change in C of over 200%.

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*Atmospheric thermal stability is defined as the difference between temperatures at elevations of 30 and 3 meters. Total moisture detention is a measure of the soil moisture content that accounts for precipitation, evapotranspiration, run-off, storage of water in the ground and the retention of additional water as snow and ice (see Cox, et al. [CBK 70] and references therein).

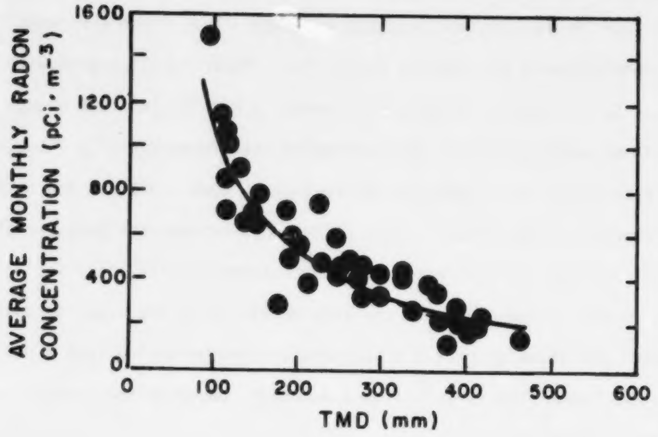

Figure 3.1. Average monthly radon concentration in ground-level air as a function of total moisture detention (TMD) in soil. Taken from Cox, et al. [CBK 70].

Auxier [Aux 73] investigated the effect of moisture content on concrete blocks. He measured radon exhalation from two blocks--one with low (2 to 4% by weight) and one with a higher (6-8%) moisture content. The moisture content was determined from the mass difference between the "moist" blocks and after baking them. The exhalation results showed a "strong dependence ..... on the moisture content of the concrete." Differences on the order of 30% were observed with higher radon exhalation resulting from higher moisture content which was obtained by placing concrete blocks in atmospheres with varying water vapor pressures.

Auxier [Aux 73] also investigated the influence of humidity on radon exhalation from concrete. He found that with increasing relative humidity from 60% to 80%, the exhalation decreased by approximately 30%. This work is the only known quantitative study of the effect of humidity on radon exhalation from building materials.

III.C.2. Pressure Effects

The dependence of radon exhalation on pressure has been noted many times [Zup 34; Kov 45; Oka 56; Ros 57; Tan 59; KSE 64; SEK 66; PoP 69; Bak 71; ClW 74; Jon 75; UN 77; Tan 78; SBU 79], and there is general agreement that the exhalation increases as underpressure increases. Several reports by Bates, et al. (see [EdB 80] and references therein) have treated exhalation from mine walls. Perhaps the only significant study of the systematics of the underpressure effect in building materials was performed by Jonassen and McLaughlin [JoM 77; McJ 78] who examined concrete. They found the radon flux density to increase linearly with underpressure as shown in figure 3.2. Their results agree with the frequently quoted value for soil: a 1% difference in barometric pressure results in a 60% change in the exhalation rate [Har 78]. It should be emphasized that the exhalation rate is essentially independent of the value for the pressure P, and that it is pressure differences between pore air and outside air, ΔP, that cause significant changes in exhalation. Such differences between the pressure within the interstitial pore spaces of a material and the external pressure can conceivably exist since the relaxation time for the pore spaces to equilibrate in pressure with the outside air is of the order of one hour (see Section I.D.7). It is not unusual for there to be a 1 or 2% pressure change during such a period of time, so that the variability in the radon flux density from a material, based on the results shown in figure 3.2, could be as much as a factor of 2.

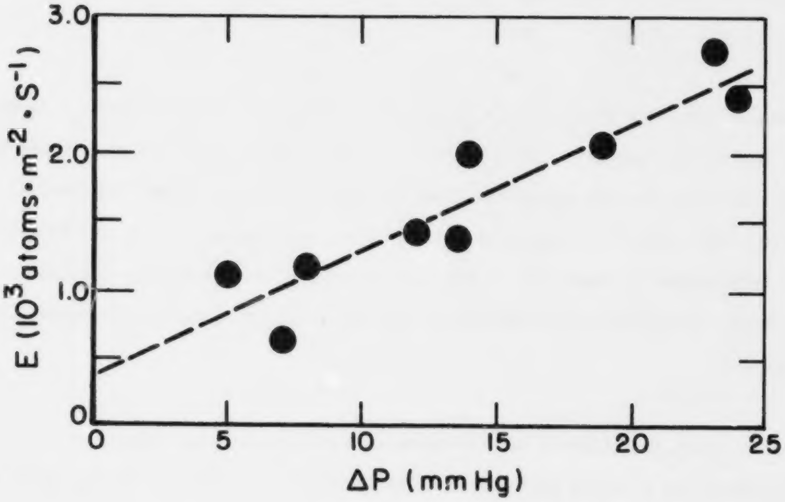

Figure 3.2. Radon flux density from a concrete as a function of the under pressure ΔP. Taken from Jonassen and McLaughlin [JoM 77].

III.C.3. Temperature Effects

The influence of temperature changes is apparently quite complex as evidenced by the substantial number of temperature-related meteorological variables found by Steinhausler [Ste 75] to be correlated with radon exhalation. Two general types of behavior in soils have been noted in the literature.

The first type is the widely observed diurnal variations [Zup 34; Kov 46; Ros 57; WiH 60; HOS 66; Kir 70; SNS 70; WCS 72] that are associated with the competition between two factors: (i) changes in convective flow due to temperature differences in the soil from day to night; and (ii) turbulent mixing in the atmosphere which leads to an increase in exhalation in the daytime and a reduction at night [UN 77].

The second type of behavior is the temperature effect arising from the direct heating of soil samples. Shashkin and Prutkina [ShP 70] report a sharp decrease in emanation power after roasting samples at 100 to 600 °C. They reported that there is a partial restoration in the emanation power after keeping the roasted samples under normal conditions or moistening them. The authors offer a number of hypotheses relating to the internal effects (e.g., radon adsorption onto internal surfaces and the effects of capillary water films) to explain the results. Similar considerations are also discussed by Tanner [Tan 78] (see also Chapter II.).

One of the more amusing temperature "effects" was observed by Okabe [Oka 56] who measured the diurnal variation of atmospheric temperature and radon concentration. The results, which are partially illustrated in figure 3.3, clearly indicate a strong correlation between temperature and radon concentration--this change in temperature coming after the change in radon concentration! It should be noted that, in this case, should a correlation analysis be performed, a strong association between the variables would be found (e.g., as indicated by the square of the correlation coefficient). This certainly does not imply that a change in the radon concentration caused a variation in the temperature, or vice versa. It is probably a good example that calls into question the validity of drawing conclusions from mere searches for associations or correlations for the purpose of identifying the variables that influence radon exhalation.

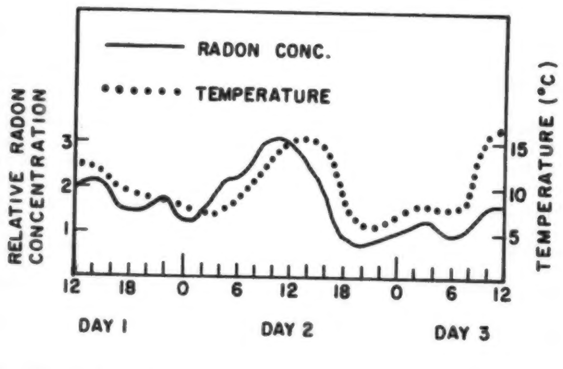

Figure 3.3. Diurnal variation of atmospheric temperature and atmospheric radon concentration. Individual data points, shown in the original from Okabe [Oka 56], have been omitted.

Only two quantitative studies of temperature effects on building materials are known, and these are contradictory. Gabrysh and Davis [GaD 55] found roughly a 20% increase in radon exhalation from a concrete sample when it was heated from 20 to 59 °C. Auxier [Aux 73], in contradistinction, concluded that there is a "negligible temperature effect on the exhalation of radon from concrete between 23 and 43 °C. Considering the number of unknown internal variables which may influence these results (e.g., moisture content or porosity of the materials), this discordance may be discomforting, but not surprising. For example, Tanner, in an unpublished study of a sample core of natural sandstone which had the appearance of a uniform sand-aggregate concrete, observed a very irregular, but steady-state decrease in radon exhalation with increasing temperature. He inferred that temperature influenced the exhalation of radon indirectly through its effect on the water distribution in the sample material [Tan 80].

III.C.4. Other Factors

Other factors that have been reported to affect radon exhalation are wind speed, seasonal changes, soil disturbances, and ground cover [Oka 56; EKS 62; KSE 64; Pea 65; PeJ 65; Pea 67; GLF 70; IsH 70; SNS 70; BaK 71; MeM 73; UN 77; UN 80]. Most of this information is qualitative in nature, however, and of little value in estimating or predicting radon exhalation. Tanner [Tan 78] also describes possible internal effects such as the role of radon adsorption onto surfaces, but these are not well understood.

III.D. Conclusions and Recommendations

Much of the data presented in the preceding three sections is summarized in tabular form in Table 3.9. The first category (A) presents ranges for typical values of radium concentrations in common building materials, and estimates the corresponding expected variation in an exhalation rate or radon flux density, E, due to the indicated natural variation of the radium content of these materials. The variation in E is given as the expected maximum value divided by the expected minimum value. It must be emphasized, however, that the radon flux density from a material may not necessarily scale proportionally with the radium concentration. The second category (B) merely tabulates the range and ratio of typical values for the radon flux density from materials. The third category (C) summarizes the known variations in E for given values or changes in the meteorological variables. In general, the variations in E, as reflected in the values of E_{max}/E_{min}, are much less affected by meteorological variables than they are by the choice of materials. It must also be emphasized that the values in Table 3.9 are only meant to be representative. They are very selective and are based on only a very limited set of results.

Based on this review, several general conclusions, and hence some recommendations, can be made.

The first conclusion is that existing data are not sufficient to quantitatively predict or estimate radon flux density or exhalation rates from building materials. Although the flux density from a material such as concrete may be estimated a priori within one or two orders of magnitude, more quantitative estimates are not possible because the exhalation process is exceedingly complex and very dependent on the specific characteristics of the material, and because many of the causal internal and external variables are only qualitatively understood.

Table 3.9

Variable	Range of Typical Values or Change in Variable	$\left(\frac{E_{max}}{E_{min}}\right)$
A. Radium Concentration		
concrete	0.6-2.0 $pCi \cdot g^{-1}$	3
concrete block	0.2-4.	20
brick	0.2-1.7	9
phosphogypsum	17-33	2
B. Radon Flux Density		
soils	0.1-1.4 $pCi \cdot m^{-2} \cdot s^{-1}$	10
concretes	0.001-0.2	200
phosphogypsum	2.-120	60
uranium mill tailings	0.8-2200	3000
C. Meteorological Effects		
moisture content	2-4% to 6-8% (by weight)	6
relative humidity	60% to 80%	1.3
pressure change	1-2%	2
temperature	23 to 43 °C	negligible [Aux 73]
temperature	20 to 59 °C	1.2 [Oka 56]

The second conclusion is that only "technologically enhanced" soil and building material exhalation rates are normally of concern as major contributors to indoor radon concentrations. If we establish a "concern level" at 10% of 3 $pCi \cdot L^{-1}$ (corresponding to 10% of 0.015 WL, assuming an equilibrium factor of 0.5), then in a single-room structure (with a height of 2.5 m and a ventilation rate of 0.5 hr^{-1}) the corresponding radon flux density from the floor surface would be approximately 0.1 $pCi \cdot m^{-2} \cdot s^{-1}$. This is above the radon flux density from most building materials (see Table 3.7). Further, if we assume that a concrete slab floor provides a 25- to 50-fold diminution in the radon flux density from the underlying soil, then the "concern level" for the radon flux density from soils is in the range of about 2 to 5 $pCi \cdot m^{-2} \cdot s^{-1}$. This is also above the radon flux density from naturally-occurring soils (see Table 3.3). If, however, this diminution is not achieved or if the radon enters the structure through cracks or openings in the foundation, then the radon flux density from soils may approach or exceed the "concern level," and may be a substantial contributor to the indoor radon.

The above considerations do not apply to buildings with very low ventilation rates, or if other air-dynamic mechanisms (e.g., the "stack effect" suggested by Nazaroff--see introduction of Chapter III) are operative. Nor do they apply to "technologically-enhanced" soils or materials. Such materials would certainly include uranium mill tailings and phosphate slags which have maximum observed radon flux densities above the minimum "levels of concern." Their incorporation into buildings or near building sites may significantly contribute to indoor radon concentrations. Hence, the third conclusion is that the use of "technologically-enhanced" raw materials in building components needs more investigation. Exhalation rates from uranium and phosphate mill tailings vary over two or three orders of magnitude with the lower end of the ranges approximating values for the more radioactive soils found in the natural environment (see Tables 3.4 and 3.5). Considering the high radium content of phosphogypsum and the wide variety of potential uses in building materials, exhalation rates from these materials should be carefully monitored. Similarly, the use of slags and fly ash as concrete fillers should be evaluated. Local variabilities in these materials may be expected to be extremely important.

The fourth conclusion is that meteorological effects due to pressure changes, temperature, humidity, and even moisture content do not seem to be of great importance individually in causing significant changes in radon exhalation. There is, however, very little quantitative information and more thorough studies of these effects may deserve attention.

The fifth conclusion is that there remain some unanswerable questions and uncertainties concerning radon exhalation from building materials and within buildings. For example, there is a large variability in the exhalation rate from soil possibly due to moisture detention which suggests that there may be corresponding large variabilities in radon exhalation from wet concrete basements. The magnitude of this effect is unknown and further investigations with water-soaked concrete blocks may be revealing. Similarly, it appears that an integral 10-cm concrete slab provides a substantial reduction in radon flux from soils, but there is no indication in the literature as to whether this is typical or that factors of 100 or even 1000 are attainable by judicious selection and treatment of materials.

CHAPTER IV

ASSESSMENT OF MEASUREMENT METHODOLOGIES

Most of what is known about radon transport through or exhalation from building materials is based upon observations and measurements in laboratory or in situ (field) settings. Therefore, an understanding of these previously employed measurement methodologies is necessary in order to evaluate the applicability and validity of existing transport and exhalation data, as well as to plan future laboratory transport and exhalation experiments.

This review considers and summarizes only those laboratory-and field-based measurement methods that are most directly applicable to studies of radon transport and exhalation from building materials. Four major areas are considered:

A. assay of gaseous radon samples;
B. laboratory measurements of radon exhalation;
C. in situ measurements of radon flux density and concentrations in soil gas and pore spaces; and
D. in situ measurements of radon concentration in indoor air.

Although other types of measurement methods (e.g., for individual radon progeny concentrations in air or working levels)* may be indirectly applicable, they are not as central or directly relevant to transport and exhalation studies. For the four areas outlined above, the various measurement methods will be briefly described; and to the extent possible will be evaluated (1) by considering their applicability for measuring the quantity of interest, (2) by identifying some of the limitations, underlying assumptions of the methods, and the most serious sources of inaccuracy, (3) by estimating their sensitivity, precision, and accuracy, and (4) by identifying some of the calibration requirements. Often, it will only be possible to identify what is unknown or undocumented. It will also be apparent in some cases, that a more critical assessment and detailed method evaluation should be performed, but this type of effort is outside the scope of this report.**

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*The wide variety of techniques used for the measurement of radon and related quantities (progeny concentrations, working levels, etc.) have been summarized by Budnitz [Bud 74], and more recently by Breslin [Bre 80] and by George [Geo 80].

**This need for critical assessments of measurement methodologies has also been recently identified by the Radiation Policy Council Task Force on Radon in Structures [RPC 80].

IV.A. Assay of Gaseous Radon Samples

The assay of gaseous radon samples is a common element in many of the methods described in Section IV.B. (laboratory measurement of radon exhalation), in Section IV.C. (*in situ* measurement of radon flux and soil-gas concentration), and in Section IV.D. (*in situ* measurement of radon concentration in indoor air). The assay methods can be subdivided into two classes: internal gas counting techniques, and scintillation counting techniques.

IV.A.1. Internal Gas Counting Techniques

For these techniques, the radon is introduced, together with a suitable counting gas, directly into a chamber or "counter" whose characteristics primarily depend on the operating voltage of the chamber (i.e., in the ionization, proportional or Geiger-Müller regions). Although a variety of shapes (parallel-plate, cylindrical, spherical) are possible, such counters usually consist of a cylindrical cathode with a wire anode (typically 0.02-0.03 mm in diameter) stretched along the axis of the cylinder. The physics and operating characteristics of these types of counters are described in a number of elementary texts and other references [Pri 64; MAG 79; NCRP 78].

Irrespective of the type of counter which is used for the radon assay, an associated gas-handling system is usually necessary. This system is used for separating and purifying the radon sample, mixing it with the counter filling gas, and transferring the gas into the counter. These systems are commonly associated with methods for assaying ^{226}Ra, and have been extensively developed for many applications by Evans [Eva 35; Eva 37], by Curtiss and Davis [CuD 43], and by Harding *et al*. [HSS 58]. Similar radon gas-handling systems have been described by Blanchard [Bla 64], by Harley [Har 72], and by Krieger and Jacobs [KrJ 78].

Ionization chambers, both direct-current and pulse (see, for example, [NCRP 78] for a discussion of these types), are the most commonly employed types of internal gas counters for the assay of radon. Any of the counters, however, are subject to certain limitations associated with the nature of the gas admitted into the counter [DaH 52]. The operation of current ionization chambers is very adversely affected by the presence of ions and condensable vapors in the gas sample. Pulse ionization chambers, proportional counters, and G-M tubes are also affected by these factors, as well as by the presence of electronegative gases (such as oxygen). These limitations necessitate the ancillary gas-handling and purification system. Proportional counters and G-M tubes are also extremely sensitive to changes in counter-gas mixture and pressure.

Internal gas counting techniques are primarily laboratory-based, and have limited applicability for in situ field measurements. Because it is less convenient than other methods, it is used for field measurements only when great sensitivity is required. Although other sample collection schemes can be employed in the field (see Sections IV.D.), normally a grab sample of air is collected with an evacuated glass flask (typically one liter volume) and then returned to the laboratory for processing and assay [ABL 77; ISC 72; Bre 80; Bud 74].

Internal gas counting techniques are very sensitive, and conceptually can be very accurate. Typical chambers have overall detection efficiencies of 85%. With one-liter air samples, sensitivities of 0.01 to 0.05 $\text{pCi} \cdot \text{L}^{-1}$ for pulse ionization chambers have been reported [Bre 80; Geo 80]. An overall accuracy (more correctly, inaccuracy) of plus or minus several percent is achievable, but may be limited in low-level samples to the imprecision ("counting error") arising from the nature of the radioactive decay process itself. In assaying radon, it is necessary to use counting intervals that are long compared to the halflife of the individual members of the ^{222}Rn subseries (^{222}Rn through ^{214}Po). Consequently, the approximations normally used for estimating the statistical "counting error" are not valid [LuW 64]. The random uncertainty associated with the radioactive decay process is given by the expression $S \simeq \sqrt{JI}$ where I is either the mean number of observed events ("counts") for counting operations, or is the product Qq of the total charge (Q) collected in an ionization chamber in the counting interval and the average charge (q) released per interacting particle, and where $J = \sigma^2/\mu$, the variance in the number of observed events from the radon subseries (i.e., ^{222}Rn parent plus progeny) divided by the mean number of observed events for the subseries. The methodology for this problem has been solved by Lucas and Woodward [LuW 64], and they have tabulated values of J for the ^{222}Rn subseries for a variety of combinations of counting interval and detection efficiency. For typical low-level samples, the average uncertainties attributable to the "counting statistics" alone may be of the order of 2-8% [Bar 75].

However, uncertainties of this magnitude can be achieved only if the gas sample is carefully and quantitatively processed and transferred to the chamber, and if the chamber is accurately calibrated. Internal ionization chambers are used chiefly for comparative measurements [NCRP 78], i.e., the assay is performed by comparing the chamber output from the unknown radon sample with that from a sample prepared in the same way and containing a known amount of radon activity. Many of the better laboratories calibrate their ionization chambers with radon sources from standard radium solutions (e.g., NBS ^{226}Ra solution Standard Reference Materials) utilizing a widely-employed "bubbler" arrangement (see, for example, [WaH 69; Har 72; KrJ 78; Geo 80; Bec 75]). The major sources of inaccuracy arise from errors in the gas-handling procedures used during the sample processing and in the chamber calibration. These gas-handling procedures and subsequent counting techniques are generally a very complicated sequence of many individual unit operations or steps. It must be recognized that virtually every step in this sequence introduces a conceivable source of inaccuracy.

IV.A.2. Scintillation Counting Techniques

For the scintillation-counting techniques, the radon sample is introduced into a suitable vessel (chamber, flask, tube, etc.) containing a silver-activated zinc sulfide phosphor which emits light that can be detected with photomultiplier tubes when the α particles from decay of the ^{222}Rn and progeny interact with the phosphor. The basic physics and operating characteristics of scintillation detection devices are also described in a number of elementary texts and other references [Pri 64; MAG 79; NCRP 78]. Two distinct techniques, one utilizing electrostatic collection, are employed.

In the electrostatic-collection technique, the initially positively-charged recoiling daughter atoms from the decay of radon are collected by means of an electric field, on to a ZnS(Ag)-coated electrode within the chamber containing the radon sample [BrM 52]. Normally, the electrode (usually a screen), may be viewed from outside the chamber by a phototube [MaS 58; RoD 66; NCRP 78], but the electrode may be removed after a suitable collection period and placed in an external α particle detector [KRH 76].

In the second technique, the gas sample is contained in a vessel whose walls are coated with a ZnS(Ag) phosphor [DaH 52; VDT 55] and viewed by one [Luc 57] or two [KSL 64] phototubes. The scintillation flask, popularly known as the Lucas cell [Luc57], is a widely employed radon assay method used both in the laboratory and in the field [Luc 57; Har 72; Bre 80]. Scintillation flasks both commercial and home-made, come in a variety of sizes (nominally 50 mL up to 650 mL), are constructed of either glass or plastic walls, and are equipped with one or two valves. The flasks are usually pre-evacuated for collection of a sample of whole air, and then mounted on the phototube assembly for counting, usually after a delay of a few hours to allow the radon decay products to come into radioactive equilibrium with the radon. For field use, the flasks may be returned to a laboratory for counting, or may be measured entirely in the field with the use of portable, battery-powered photomultiplier assemblies [Bre 80]. The DOE Environmental Measurements Laboratory has also developed a continuous scintillation flask monitor in which air is continuously drawn through the flask (see Section IV.D.3.) [HLW 57; CoT 77; Tho 77b; Geo 76; Bre 80].

Unlike internal gas counting techniques, scintillation flasks are relatively insensitive to the gas mixture and the presence of ions, dust, or condensable vapors [DaH 52; Luc 57]. In the laboratory, however, they can be and frequently are employed with the same type of gas-handling systems which are used for separating, purifying, and transferring radon samples.

In routine use, the sensitivity of scintillation counting techniques is about one-tenth that for the internal gas counting techniques. The sensitivity may be increased by a factor of two to four by using larger flasks and increasing the counting interval [Luc 57; Geo 76; Bre 80], or by greater factors by preconcentrating the samples prior to filling the scintillation cells. The sensitivity is often controlled by the background counting rate of the scintillation vessel and phosphor. Although early scintillation tubes were reported to have backgrounds as large as 5 alphas per minute primarily due to the phosphor [DaH 52], the background counting rates for most scintillation flask assemblies are now typically 0.1 counts per minute [Luc 57; Geo 76].

The accuracy considerations and calibration requirements are similar to those described for the internal gas counting techniques. Perhaps because scintillation flasks are commercially available and more widely employed, improper flask calibrations are more frequently encountered. George [Geo 80] reported that in an interlaboratory calibration exercise in which 12 groups took part, the reported values of half of the participants (6 of them) deviated by more than 20% from the expected value. Every scintillation flask must be individually calibrated and must be used with the same counting equipment with which it was calibrated. Flasks counted on different equipment can be substantially in error because of different operating characteristics of the photomultiplier tubes [Geo 80]. Other frequently encountered sources of inaccuracy are diffusion of the radon into the phosphor and walls of the flask, and leakage from the flask [Geo 76; Geo 80]. Internal field effects occurring with electrically-insulating internal chamber surfaces may result in a consequent variation in the detection response due to the migration of radon progeny. Lucas [Luc 57] reported that with an insulating window, trace impurities produced localized internal fields which created a variation in the distribution of the radon progeny within the flask. Kraner, et al. [KSL64] also observed inconsistent counting rates due to the presence of an electrostatic field within the flask that was induced by the applied voltage. Lucas corrected the variations by making the transparent internal surface of the flask window electrically conducting. Kraner eliminated the problem by providing grounded external electrodes and electrostatic fields. The Environmental Measurement Laboratory, and other laboratories, have eliminated effects of this type by coating all of the internal surfaces of the flask with ZnS(Ag) phosphor and by grounding the cathode of the photomultiplier tube [Geo 76].

IV.B. Laboratory Measurement of Radon Exhalation

The literature on laboratory measurements of exhalation of radon from materials is extensive and dates back to the 1930s (see, for example, [Hah 36; Hai 64]). Most of this work has been of a microscopic nature such as to characterize the emanating power of crystals, minerals and rocks, or used to study the adsorption characteristics and surface properties of materials. These were mainly performed for either geological interest or for various physico-chemical applications. Studies specifically related to the measurement of radon exhalation from macroscopic quantities of building materials, however, are quite limited. The measurement methods used in these studies can, in large part, be characterized as follows:

i) All of the studies are similar in principle and are based on the same general methodology.

ii) The specific apparatus and detailed operating procedures for each study are unique.

The general methodology consists of placing the sample of building material into a sealed vessel which is operated under controlled conditions; allowing the radon to escape (exhale) from the sample and build up inside the vessel; collecting and/or sampling and transferring the radon in the vessel atmosphere; and assaying the radon.

Many different types and sizes of vessels to contain the samples have been used. These include various laboratory "de-emanation" flasks [Bar 73; Bar 75], chambers [Naz 79], and vacuum-tight desiccators [PeS 78]. In an early study of the effect of radiant heating on radon exhalation from concrete, Gabrysh and Davis [GaD 55] used a simple glass tube (approx. 5 cm diam. x 30 cm) molded around a smaller heating tube. Jonassen and McLaughlin [JoM 76; JoM 78] used two different types of containers: a 200 L cast iron pressure chamber, and simple sheet metal barrels with an internal volume of approximately 120 L. Similar types of metal cylinders were also used by Culot, et al. [COS 73; COS 76]. Auxier and coworkers [Aux 73; ASK 74] employed a large chamber constructed of polyethylene supported by a wood frame.

To a large extent, the degree to which the experimental conditions in the vessel are controlled determines the scope of the inferences that may be drawn from the measurement results obtained. In many reports, the influencing variable under study (e.g., temperature or pressure) is controlled, but no mention is made of the control placed on other factors that may affect the results. Therefore, the results obtained may be of only limited validity.

The length of time that the radon is allowed to build up within the vessel may range from a few hours or less [JoM 76] to many days [GaD 55; Bar 75]. It depends mainly on the exhalation rate of the material under study and on the sensitivity of the sampling and assay procedure that is utilized.

One of the more common operating procedures consists of performing a single sampling and assaying of the radon in the vessel at the end of the buildup period. The exhalation rate is then determined from the total amount of radon in the vessel (or from an aliquant of the total) and the buildup time interval. Frequently, the vessel is purged with radon-free air or other gases prior to sealing to remove any initial radon from the vessel's atmosphere. This type of procedure suffers in that only a final determination of the amount of radon released from the material is made, and any potential problems (e.g., loss of radon by leakage from the vessel, or "back diffusion" into the sample) will be masked.

An alternative technique is to study the buildup of radon in the vessel over time by either continuous monitoring or periodic sampling [JoM 76; JoM 78]. This procedure is similar in principle to the accumulation method (see Section IV.C) used to determine radon flux density or the exhalation of radon from soils [KSE 64; Wil 74; WCS 72; BJK 75]. The exhalation is determined from the initial slope of the growth curve, and in this way obviates, to an extent, the possible problems of leakage and back diffusion [BJK 75; JoM 76; JoM 78; Jon 80].

Possible variations in the sampling and collecting procedures include collecting the radon in the vessel at the end of the buildup period by gas-sweeping [Naz 79], vacuum-transfer (see, for example, [PeS 78]) and volume-sharing transfer [Bar 71; Bar 75] techniques, or by periodic sampling of only a small portion of the vessel atmosphere with an evacuated flask (perhaps directly into the chamber used to assay the radon) [JoM 76]. Obviously, in order to measure only the naturally escaping fraction of radon produced within the sample material, the material should be disturbed as little as possible during sampling and collecting. Therefore, the volume-sharing transfer method is less susceptible to this possible problem than the gas-sweeping and vacuum-transfer techniques. The grab sampling techniques introduce periodic underpressures in the vessel which may perturb the exhalation rate, and may result in inhomogeneous samples that are not truly representative of the radon concentration in the entire vessel. Jonassen has reported that, through a variety of experiments, it appears that these two possible problems are not significant [Jon 80]. The radon sample from the vessel may also be filtered to exclude the radon-progeny radionuclides from the samples (see, for example, [PeS 78]). Further variations include collecting the radon in a charcoal trap [Naz 79] or in a low-temperature condensation flask [JoM 76] for subsequent preconcentration and/or purification of the radon sample (see Section IV.A). Auxier and coworkers [Aux 73; ASK 74] used a closed-loop flow system to collect the radon progeny on filters which were subsequently alpha counted with surface-barrier detectors. Although they utilized a vibrator inside their vessel in an attempt to ensure a constant distribution of dust particles, it is unclear how the concentration of condensation nuclei was controlled, or how they accounted for plate out of the radon-progeny radionuclides.

In laboratory measurements of radon exhalation, the radon samples are most frequently assayed by scintillation counting or internal gas-counting techniques (see Section IV.A). The samples are either introduced directly into the counting chambers or purified with an associated gas-handling system prior to assay. The latter is not as prevalent a practice. Although several of the studies [JoM 76; Bar 73; COS 73] include a description of the calibration procedures which were used for the radon assay (it is important to include the radon collection efficiency), the number of studies in which they are not reported is dismaying. One can only conjecture whether they failed to report the calibration procedure, or failed to calibrate!

As indicated, the reported methods of laboratory measurement of radon exhalation from building materials all utilize the same general methodology, but offer a nearly semi-infinite variety of possible variations in the specific apparatus and operating procedures. Because each study is somewhat unique to itself, it is difficult, if not impossible, to unequivocally evaluate and state the adequacy and reliability of any one of these possible sets of variations. One can, however, summarize the most common failings or possible sources of inaccuracy which may occur. Most of these have been mentioned, and they include: (1) uncontrolled experimental conditions; (2) unaccountable losses or leakage of radon from the vessel; (3) "back diffusion" of radon into the sample material; (4) adsorption of radon onto surfaces in the vessel or inhomogeneous radon concentrations in the vessel resulting in unrepresentative samples; (5) perturbations of the exhalation rate during sampling and collection; (6) sampling and collection losses such as adsorption of the radon in the transfer system; and (7) errors in the assay and calibration procedures.

IV.C. In Situ Measurement of Radon Flux Density and Concentration in Soil Gas and Pore Spaces

In situ measurements of radon flux density and radon concentrations in soil gas and in pore spaces are the two classes of measurements that are most applicable to radon transport and exhalation studies of building materials. Measurements of radon flux density, for example, are often used to evaluate the sources and pathways of radon into a building [DSM 78; GHA 80]. Similarly, soil gas concentrations of radon are useful in evaluating the diffusion and transport of radon through soils near building foundations [Sco 80], and for determining diffusion coefficients through building materials (e.g., building foundations). Scott [Sco 80], however, has cautioned that although measurements of radon flux density and soil gas concentration should be valuable in determining radon transport in soils, the large variability seen in both of these measurements leads one to suspect that the radon concentration in the upper soil layer is so greatly influenced by atmospheric effects that short-term measurements may not be very helpful. Similarly, in those porous soils where radon transport may be significant, it may be very important to not disturb the surface pressure distribution, or else the radon concentration near the surface may be changed by the presence of the measuring equipment.

IV.C.1. Radon Flux Density

Radon flux density is just the exhalation rate of radon from a given surface area. Although most of the methods for measuring radon flux density were developed for studying radon exhalation from the earth's surface (see, for example, [WCS 72]), the methods may, with slight variation, be applied to measure the flux density from any type of surface. Six main categories of methods can be recognized:

(i) Accumulation Method
(ii) Induction Method
(iii) Flow Method
(iv) Adsorption Method
(v) Vertical Profile Method
(vi) Soil Concentration Gradient Method

The accumulation method (i) involves the direct accumulation of radon in an open-faced vessel which is inverted and placed (and possibly sealed) on the surface. In this way, the vessel and surface forms a closed container for the accumulation of exhaled radon. On soil surfaces, the vessel is normally embedded several centimeters into the ground [Cul 46; KSE 64], or sealed to the ground with, for example, wetted bentonite [BJK 75]. On other surfaces, such as building materials, the vessels are sealed with epoxy resins or other caulking agents [JoM 76; JoM 78]. Many different types and sizes of accumulators have been used including large barrel drums [KSE 64; BJK 75], large surface area parallelepipeds [GLF70] and disc cylinders [WiH 60].

Although early deployments of the method [Zup 34] relied on a long accumulation period followed by a single sampling and assay of the radon, the method is now commonly performed by periodic sampling and assaying to follow the growth of radon concentration in the accumulator [KSE 64; WCS 72; BJK 75; JoM 76]. This is done because with materials of high porosity, such as most soils, the radon concentration in the accumulator will rapidly become comparable to the concentration of radon in the pores of the soil. "Back diffusion" of radon into the pores will then occur, resulting in marked departures from the growth curve which is governed by the simple radioactive growth equation. The exhalation rate is determined from the initial slope of the observed radon concentration growth curve using a well-known and developed methodology [KSE 64; WCS 72; BJK 75; JoM 78].

Wilkening, et al. [WCS 72] reported that the accumulation method, as commonly employed, is based on several assumptions: (1) the accumulation time is short compared to the radon halflife (radioactive decay corrections can, of course, be made); (2) the concentration of radon in the accumulator is significantly below the soil gas concentration to prevent appreciable back diffusion; and (3) the presence of the accumulator does not seriously perturb the exhalation process. With respect to the second assumption, Wilkening, et al. [WCS 72] recommends, as noted by Kraner, et al. [KSE 64], that the accumulated radon concentration be kept below 10% of the soil gas concentration at a depth of 13 cm. The third assumption is more troublesome and more difficult to verify. Other than by, perhaps, comparing the results obtained with the accumulation method with that obtained by other methods (e.g., the flow method)--under identical conditions--it is not clear how the third assumption can be validated.

A number of efforts have been made to control the conditions within the accumulator. This includes the use of fans to mix the accumulator atmosphere and insure homogeneous sampling [Cul 46; WiH 60; KSE 64; WCS 72; BJK 75], maintaining constant pressures in the accumulator by drawing air in during sampling [Cul 46] or using pressure-equalizing orifices [WCS 72], and monitoring and controlling internal temperatures [BJK75].

Sampling and assay of the radon from the accumulator is similar to the techniques used for laboratory measurement of radon exhalation (see Section IV.A). The techniques employed, however, are usually those more compatible to the demands of field deployment. As a result, the most common variation is the collection of the radon grab samples directly into evacuated scintillation flasks [BJK 75; WCS 72; Cle 74], although other sampling, transfer, collection and gas-counting techniques have been employed [Zup 34; Cul 46; WiH 60; KSE 64].

The most extensive evaluation and verification of the accumulation method using large approximately 100 L and 200 L barrel accumulators and scintillation-counting procedures was performed by Bernhardt, et al. [BJK 75]. They reported that the "counting errors" for the radon assays were generally less than 5% for each sample, and that the overall precision in the exhalation rate estimated from replicate measurements (both sequential measurements using one accumulator, and measurements using accumulators of different sizes) was within the estimated uncertainty of the growth curve regression analyses. The precision (actually imprecision)was typically 20% for radon fluxes of approximately 10 $\mathrm{fCi \cdot cm^{-2} \cdot s^{-1}}$, and increased to 50-100% for radon fluxes of less than 1 $\mathrm{fCi \cdot cm^{-2} \cdot s^{-1}}$. They also reported that the use of a common calibration factor for the scintillation flasks (rather than individual calibrations for each flask) introduced on additional analytical uncertainty of 6%.

The accumulation method has also been modified and used for measuring the exhalation of thoron and thoron flux density [Cro 69; GLF 70].

The induction method (ii) may be viewed as a specialized form of the accumulator method (i) which utilizes an electrostatic-collection technique. Although various forms of the induction method have been employed since the early 1900s, Rosen [Ros 56; Ros 57] is primarily responsible for adapting the method to the measurement of radon exhalation from the earth. For this method, a large surface area accumulator, containing a negatively-charged collector wire, is placed over the ground. The method utilizes the fact that some of the radioactive decay products of radon when first formed are positively charged, and these ions are collected by means of the electric field about the collector wire. The radioactive deposit on the collector is then assayed. Rosen [Ros 56] used a relatively large metal accumulator tank (approx. 3m x 1m x 1m hgt.) in which the radon decay products were measured by an internally located gauze cylindrical ionization chamber situated around the wire. Other than Rosen's work [Ros 56; Ros 57], the induction method has not been widely employed and therefore is only of limited interest.

The flow method (iii) for measuring radon flux density was principally developed by Pearson and coworkers [Pea 65; PeJ 65; Pea 67] in an effort to more nearly simulate natural conditions at the sampling site. The method uses a long, shallow collector in which the exhaled atoms of radon are entrained in a closed-loop stream of air flowing across the ground-air interface. The entrained radon in the air stream is continuously collected in a dry-ice cooled charcoal trap after first passing through drying tubes and water traps. A by-pass flow route containing another charcoal trap is usually provided to flush radon from the system prior to sample collection.

Pearson [Pea 67] outlined the principal requirements for the collecting device: (1) it should minimally disturb the soil during the placement and operation of the collector; (2) it should be constructed so that it can be sealed to the soil surface and thereby prevent both leakage from the collector and admission of external air; (3) the flow within the collector should simulate natural air movements on the free, unobstructed ground-air interface; (4) it should have a well-defined collection area in order to determine the flux density (exhalation rate per unit area); and (5) it should be durable, inexpensive and easily fabricated. The collector Pearson, et al. [Pea 67; PeJ 65] designed was a straight tunnel which permitted a reasonably constant flow across the entire surface being sampled. They found that a collection flow rate of approximately 4 liters per minute approximates average wind speed and gustiness, although the flow can be adjusted at the sampling site to duplicate, as nearly as possible, the meteorological conditions over the area being sampled.

In Pearson's work, the charcoal traps containing the radon samples were returned to the laboratory where the radon samples were desorbed by heating, flushed and transferred into scintillation counting flasks for assay (see Section IV.A). They have made a rather comprehensive evaluation of the method, including various assumptions and some possible sources of inaccuracy [Pea 65]. In addition, they reported a coefficient of variation (standard deviation divided by the mean) of 30% for a set of 8 replicate determinations of the radon flux (taken close together and within a 1-hour interval) [Pea 67].

The flow method has also been used by Israel, et al. [IHL 68] in which an ionization chamber (instead of charcoal traps and scintillation counting) was incorporated into the flow system. A similar system using a Pearson collector and ionization chamber was adopted by Styro, et al. [SNS 70] to measure thoron exhalation.

A portable and automatic instrument which directly measures radon exhalation from a mine wall has also been developed [Gre 74]. This instrument, which is based on the flow method, consists of a metal collecting cylinder (with a surface area of approximately 0.1 m^2) that is attached to the surface of a mine wall. A stream of carbon dioxide inside the collector is used to continuously sweep the mine wall, and the exit stream is filtered to remove radon progeny radionuclides. Alpha particles from the decay of the radon entrained in the exit stream are detected with an external, flow-through gas proportional counter. A prototype of the instrument was tested, and the efficiency of the proportional counter for radon in the exit stream was calibrated against an ionization chamber. The sensitivity of the instrument has not been reported, and it may not be sufficiently sensitive for most non-mine applications because of the low detection efficiency for the external proportional counter.

In the adsorption method (iv), the surface to be measured is covered directly with granular activated charcoal, and the exhaled radon is adsorbed onto the charcoal and collected. The quantity of radon collected is then determined by directly assaying the charcoal for the radon progeny radionuclides by gamma-ray spectrometry .

Megumi and Mamuro [MeM 72] used the adsorption method to determine the exhalation rate of both radon and thoron from soils. In their work, they used plastic frames 35 x 70 cm in area and 1.5 cm high to contain the charcoal. The soil surface surrounded by the frame was first covered with a sheet of gauze, then with a bed of charcoal uniformly distributed over the gauze, and sealed with a polyvinyl chloride film and a 5-cm thick sheet of polystyrene foam. Usually, they simultaneously employed four frames, each containing approximately 210 grams of charcoal, for collection periods of 5 hours. The charcoal was then removed from the four frames and sealed in polyvinyl chloride cases which were 17 cm in diameter and 8 cm high. Interestingly, the authors noted that they used a "windscreen" to prevent the gases adsorbed on the charcoal from being carried away by the wind!

The sample cases containing the charcoal were then returned to the laboratory and measured with a 5 x 5-inch NaI(Tl) scintillation detector. Thoron and radon were determined from measurement of the 583-keV γ-rays of ^{208}Tl, and 609-keV γ-rays of ^{214}Bi, respectively. Because the 583- and 609-keV photopeaks in the γ-ray spectrum are not completely resolved, the ^{214}Bi (and hence radon) determination is not made until after the ^{208}Tl has sufficiently decayed. The assay procedure was standardized with a calibration source prepared by mixing a standard radon solution with charcoal and sealing the mixture in a similar sample case.

In separate experiments using a laboratory chamber, Megumi and Mamuro [MeM 72] evaluated the radon adsorption efficiency as a function of temperature and humidity. They found that in the temperature range of 4-35 °C, the adsorption efficiency was 85-90%. The efficiency gradually decreased as the temperature increased until it reached 60% at 70 °C. The adsorption efficiency was independent of humidity in the relative humidity range of 40-90%. They also evaluated the amount of charcoal needed to effectively adsorb the radon and thoron exhaling from a given surface area. Using frames of seven different sizes, each containing the same amount of charcoal (100 g), they found that the amount of adsorbed thoron was proportional to the frame area for areas between 500 and 1500 cm^2, and concluded that more than 0.067 $g \cdot cm^{-2}$ of charcoal should be used. They postulated that the proportionality above 1600 cm^2 was lost because of "the decrease in the probability that thoron will be adsorbed by the charcoal before it decays." This explanation does not seem plausible, however, since one might expect the ^{216}Po (0.15 s) daughter or ^{212}Pb (10.6 hr) granddaughter to be more readily adsorbed than the thoron.

A more widely used variation of the adsorption method is the charcoal canister technique developed by Countess [Cou 76; Cou 77a; Cou 77b]. It has been used by many others to determine the radon flux density from a variety of surfaces both indoors and outdoors, including building materials, soils, and phosphate gypsum and uranium mill tailings piles. The technique has similarities to other charcoal adsorption methods for collecting radon, but as noted by Countess [Cou 77b], it avoids the possible radon losses and problems inherent in either transferring the radon from the charcoal as in the Pearson flow method [Pea 67] or transferring the charcoal as in the Megumi and Mamuro [MeM 72] method.

Countess used canisters from conventional personal respirators (U.S. Army M11 gas canister) which were modified by removing the metal casing from one face. The canisters have a cross-sectional area of 87 cm^2, are 6 cm high, and contain approximately 150 g of activated charcoal. They are sealed to the soil surface by simply embedding the rim of an outer protective plastic casing into the surface of the soil to a depth of about 1 cm, or sealed to other surfaces with either a clay-like material or pliable weather stripping materials. A canister is normally deployed for a period of one day or longer, and returned to the laboratory for analysis.

Similar to the method used by Megumi and Mamuro [MeM 72], the adsorbed radon in the canister is assayed with a gamma-ray spectrometer. Countess used an 8 x 8-cm NaI(Tl) scintillation detector. The exposed side of the canister was placed on the detector, and counted after the early progeny radionuclides reached radioactive equilibrium with the adsorbed radon (about 5 hours after the end of the exposure period). Analysis is performed by measuring either three or four γ rays from ^{214}Pb and ^{214}Bi (242-, 294-, 352- and 609-keV). In his first work, Countess calibrated the method by drawing measured volumes of filtered air having a known radon concentration (determined by the two-filter method, see Section IV.D.1) through the canisters [Cou 76]. Subsequent calibrations, however, were performed by direct comparisons with the more traditional accumulation method [Cou 77a; Cou 77b]. Canisters and accumulators were exposed simultaneously to several test surfaces (and positions were alternated to average spatial and temporal variations in radon flux), and used to obtain a calibration factor for the canisters. The radon flux and hence the amount of radon adsorbed by the canisters was calculated from the accumulator measurements. The calibration factor was found to be independent of exposure time for periods of several hours to several days [Cou 77b].

Countess investigated the adsorption efficiency, and reported that the canisters quantitatively adsorbed radon [Cou 77a]. Possible losses of radon from the canisters due to temperature, reduced pressure, and humidity effects during exposure and transit were also evaluated. There were losses in all cases, but for normal temperature and pressure ranges the losses were usually less than 1%. The humidity dependence was much greater, but Countess concluded that "canisters exposed to wet soil for a period of several days will not adsorb enough water to reduce the capacity of the canisters for radon by more than 10-15%" [Cou 77a].

By making multiple measurements on several test surfaces in which the canisters were deployed simultaneously in a relatively small area, Countess (Cou 77a) reported that the precision of the canister method ranged from 6% (coefficient of variation) for an outdoor location in the high radon flux density phosphate region of Florida (10 $pCi \cdot m^{-2} \cdot s^{-1}$) to 15% for a measurement on soil in New Jersey (1 $pCi \cdot m^{-2} \cdot s^{-1}$). Wide variations in radon flux obtained by the charcoal canister method have sometimes been seen over even small areas. A two-laboratory intercomparison study was recently performed [Hor 79] to determine if the actual analysis of the canister is the major contributor to this wide variation. Fifty canisters exposed to radon flux densities ranging from approximately 2×10^{-16} to over 10^{-14} $Ci \cdot m^{-2} \cdot s^{-1}$ were assayed in one laboratory and then shipped to and assayed in the second laboratory. The two assays on individual canisters typically differed by 10 to 30%, but the 50-canister average difference between the two assays was 16%.

The charcoal canister method is not as sensitive as some of the other radon flux density methods. Countess reported a typical lower limit of detection of 70 pCi which corresponds to the radon accumulated by a canister in a four-day exposure at a flux density of 0.03 $pCi \cdot m^{-2} \cdot s^{-1}$ [Cou 77a]. To achieve adequate sensitivity, the canisters usually must be deployed for at least a day or longer. As a result, the canister method provides only an average (time-integrated) flux density rather than the series of more "instantaneous" values which are possible by the accumulator method.

The charcoal canister method is probably now the most widely used method for determining radon flux density. In large part, this is because it is relatively simple, inexpensive, and much less labor intensive than the other methods. Several types of commercial canisters have been used for a variety of purposes by many different groups. The method has been used indoors to locate and measure radon sources and routes of entry into buildings. Various adapters have been employed for mounting canisters in corners, across wall-wall or wall-floor joints and over cracks in concrete foundations [Cou 77a; Sco 80]. The original development and employment of the method by Countess, and subsequently continued by the DOE Environmental Measurements Laboratory [Bre 80], has been admirable. Unfortunately, not all users of this method are as careful in performing the method, evaluating their use of it, and calibrating it. On inquiry with several users, it has been learned that some merely use Countess' original calibration factor, and that substantial losses of radon from the canister are accounted for by using the concept of an "effective halflife" (which assumes, of course, that the desorption and loss of radon from the canister over the entire collection, transit and assay time periods is a first-order kinetic process- which is not likely).

It is not clear to what extent the adsorption method, by the direct placement of the charcoal on the exhaling surface, perturbs the exhalation rate. Of the radon flux density methods, it probably most poorly simulates natural conditions. Furthermore, perhaps there should be some concern over the relatively small area sampled by charcoal canisters. Since the popular adsorption method using charcoal canisters is primarily calibrated against the accumulation method, both of these effects will be partially masked, and an independent assessment of them (e.g., by comparing the adsorption method to other methods) is not possible.

The vertical profile (v) and soil concentration gradient (vi) methods are indirect methods whereby an average radon flux density is inferred from measurements of the vertical concentration gradients of radon in either the atmosphere or the soil. Both methods are based on a large number of assumptions, and therefore have limited utility for the direct measurement of radon flux density from surfaces.

In the vertical profile method, the vertical distribution of radon in the atmosphere is determined by measuring the radon concentration at ground level through the mixing layers by means of captive balloons or aircraft [Kir 70; WCS 72]. The radon flux density from the soil is then estimated by summing the vertical radon concentration distribution (taking into account the structure of the atmosphere). The method neglects advective effects (localized changes in the radon concentration gradient caused by mixing and air movements in the gradient), and is based on the assumption that the total radon content of a vertical column of air (extending into the atmosphere) must balance with the radon flux into the base of that column.

The soil concentration gradient method is based on assuming a particular transport model (see Section I.D.). Radon flux density and soil gas concentration are known to be correlated [WCS 72]. With a given transport model, the radon flux density at the surface of a material can be related to the profile of the concentration gradient of radon in the interstitial pore volume [ClW 74; JoM 77]. This method was used by Senko [Sen 68] to estimate radon flux density from soils (and an effective diffusion coefficient) by measuring soil radon concentrations at two depths (see Section IV.C.2) and assuming that the radon concentration in the soil was exponentially distributed with depth. More recently, Mochizuki and Sekikawa (MoS 78) using silicon surface-barrier detectors measured the concentration of the radon daughter ^{218}Po in soil air at various depths to infer radon flux density. Inasmuch as the measurement of radon flux density from materials may actually be used to validate models of radon transport in the materials, a method that is based on an a priori transport model assumption cannot be used.

Very few efforts have been made to directly intercompare the various methods for measuring radon flux density. As indicated earlier, the widely used adsorption method (iv) using charcoal canisters is commonly calibrated against the accumulation method (i). Wilkening, et al. [WCS 72] reviewed various measurements by the accumulation (i), flow (iii), direct charcoal adsorption (iv) and vertical profile (v) methods. On comparing the accumulation, flow and vertical profile methods, they concluded that the accumulation and flow methods generally agree (usually within 30%), but that the vertical profile measurements "tend to yield higher values when compared at the same location" (WCS 72). They also noted that the vertical profile method (unlike the accumulation and flow methods) has the advantage of not disturbing the surface and of integrating over large areas, but that its assumption of no advective flow is extremely restrictive and exceedingly difficult to correct for. In contradistinction, Senko [Sen 68] compared the accumulation (i), Pearson flow (iii) and soil concentration gradient (vi) methods and reported that radon flux densities measured by the flow method averaged 3.2 times higher than values obtained by the accumulation method. He stated that the "reasons for this discrepancy are not clear" [Sen 68]. He also found that the soil concentration gradient method (with its exponential radon concentration profile assumption) was in better agreement with the flow method under "convective conditions", but was closer to the accumulation method under "inversion conditions" [Sen 68]. Clements and Wilkening [ClW 74] used a more sophisticated transport model to compare the accumulation and concentration gradient methods. (The experiment was performed, of course, to test the radon-transport model.)

In a laboratory experiment using a column of radium-bearing porous material in which they could induce interstitial gas flow, they compared the direct measurement of radon flux density by the accumulation method to a predicted flux density calculated from the measurement of the radon concentration profile in the column. The agreement was exceptionally good.

Some of the above discrepancies may, qualitatively at least, be rationalized. The vertical profile method (which is only useful for geographical surveying of large areas) may give higher values for the radon flux density since its use does not disturb the surface. The extent to which the actual measurement of the radon flux density perturbs the flux density itself is, of course, a serious concern. Similarly, one may expect the flow method to give higher values than the accumulation method because the former attempts to simulate natural air movements on the unobstructed surface. This type of argument may also explain Senko's [Sen 68] finding that the flow and concentration gradient methods better agreed under maximum convective flow, but not under inversion conditions.

Wilkening, et al. [WCS 72] have noted that "wide variations occur in radon flux (density) measured at different sites, at different times at a given site, and by different methods employed simultaneously at the same site." One would only hope that the differences in the method of measurement could be eliminated, so that the real spatial and temporal variations could be better understood.

IV.C.2. Radon Concentration in Soil Gas and Pore Spaces

The measurement of radon concentration in the pore space of soils has been widely used for uranium exploration [IAEA 76], as well as for understanding the transport of radon through porous materials [ClW 74; JoM 76; JoM 78].

Traditionally, sampling holes are prepared by drilling or driving various types of pipes, rods, cylinders and sampling tubes into the ground which are then covered [BoW 32; Tan 59; NMS 52; ElO 60; Sco 80]. The holes are typically 1 to 10 cm in diameter, and precautions are normally taken to prevent compacting the soil. The soil gas in the sampling hole is then sampled and assayed for radon. Sampling may be performed by vacuum-collecting, aspirating, or pumping the gas into collection systems or directly into scintillation or ionization chambers [BoW 32; NMS 52; Tan 59; SlO 60]. Variations on this theme include flow-through and recirculation systems [IAEA 76], and several techniques involve the insertion of the detection devices directly into the hole [Dyc 69; MoS 78].

The basic premise of the method is that the concentration of radon in the sampling hole equilibrates with the concentration in the interstitial pores, and that the meteorological influences on the radon concentration are the same in the pores as in the sampling hole [Kov 45; Kov 46; Tan 59]. Scott [Sco 80] has noted that unless the soil gas is drawn into the sampling tube by suction, then it is unlikely that the radon concentration in the hole equilibrates with that in the soil gas. He has reported that measurements of radon concentration in open holes are often ten times less than soil gas concentrations measured by suction.

The only known measurements of the radon concentration in the pore volume of a building material were made by Jonassen and McLaughlin [JoM 77; JoM 78]. They drilled narrow bore holes to different depths in a 30-cm thick concrete wall to obtain a pore air radon concentration profile. Sampling tubes were sealed in the holes using an epoxy resin, and the wall surface surrounding each sampling tube was also sealed with epoxy. Samples were vacuum collected from the sampling tubes using one liter evacuated flasks, and assayed by scintillation counting techniques (see Section IV.A.2). The effect of the epoxy resin on perturbing the exhalation of radon into the pore air is unknown. On the basis of leak testing the sampling procedure, they concluded that the major part of the sampled air came from wall pores, although they acknowledged that "the air in any sample comes from an undefined volume of pores surrounding the inner end of the sampling tube" [JoM 77].

IV.D. In Situ Measurement of Radon Concentration in Indoor Air

The methods used to measure indoor radon concentrations may be subdivided, as done of Breslin [Bre 80], into three groups:

1. "Instantaneous" radon concentration methods in which an air sample is obtained nearly instantaneously or over only a few minute sampling period;
2. Time-integral radon concentrations methods which provide a single, average concentration for an extended period of time ranging from a few days to a week or longer; and
3. Continuous radon monitoring methods.

The methods within each group may be further categorized as shown in Table 3.1.

Table 3.1

Methods for Measuring Radon Concentration in Indoor Air

INSTANTANEOUS METHODS

- grab-sample collection/laboratory assay techniques
- grab-sample/scintillation flask techniques (Lucas)
- two-filter tube method

TIME INTEGRAL METHODS

- large-volume sample collection/laboratory assay techniques
- TLD-based diffusion-electrostatic monitor ("PERM")
- track-etch techniques

CONTINUOUS METHODS

- flow-through scintillation-cell monitor
- flow-through ionization chambers
- automated two-filter tube method
- diffusion-electrostatic techniques (Wrenn)

IV.D.1. Instantaneous Methods

Some of the "instantaneous" sampling/laboratory assay techniques for radon concentration were briefly described in Section IV.A. Normally, a grab sample of air is collected with an evacuated flask and returned to the laboratory for processing and assay by either internal gas-counting or scintillation-counting techniques. Alternatively, the air sample may be collected directly in an evacuated scintillation flask (Lucas cell).

Another important technique for the instantaneous measurement of radon concentrations in indoor air is by the two-filter tube method [Fon 64; Jac 65; TaL 66; ThL 70]. The method consists of drawing air through a long cylindrical tube fitted with filters on both ends. The front filter removes all particulates including radon progeny radionuclides from the air. Some of the radon entrained in the air stream within the tube decays in-flight, and a fraction of the ^{218}Po daughter atoms that are produced are collected on the second filter. Typical sampling periods range from 5 to 10 minutes. Immediately following the sampling period, the second filter is removed and counted for several minutes with a portable alpha-particle detector. In addition to the tube, field measurements require a portable alpha-particle counter and an air sampling pump.

Although the radon concentration can be calculated from a simple equation that considers the tube dimensions, sampling volume, and sampling and counting time intervals, it must include a correction factor for the loss of ^{218}Po in the tube by plate out and diffusion [ThL 70]. A direct calibration, such as performed by Thomas and LeClare [ThL 70], with a known radon concentration is preferable, but this is not always possible for all users. In the absence of a direct calibration, at a minimum, the α-particle detector and ancillary electronics should be calibrated with an α-particle emission-rate standard that has the same geometric configuration as the filter. The standard should also match, as closely as possible, the ^{218}Po α-particle energy (6 MeV) and the typical emission rates of the filters. The sampling flow rate should be metered, and it also requires calibration. A correction for the self-absorption of the ^{218}Po α-particles in the filter may also be necessary.

In a laboratory experiment based on 17 replicate determinations at a radon concentration of 570 $\text{pCi}\cdot\text{L}^{-1}$, Thomas and LeClare [ThL 70] reported a precision of approximately 1.4% (coefficient of variation). If the precision scales with the inverse square root of the radon concentration, one might expect coefficients of variation of approximately 11% at 10 $\text{pCi}\cdot\text{L}^{-1}$ and 35% at 1 $\text{pCi}\cdot\text{L}^{-1}$ (under identical conditions).

George [Geo 80] identified several common sources of inaccuracy in the two-filter method. Air leakage around the front filter will increase the radioactivity on the second filter and result in an overestimate of the radon concentration. This may be avoided by pushing rather than pulling air through the tube. The front filter should be essentially 100% efficient for removing radon progeny radionuclides; and the second filter should have surface characteristics favorable for the collection and assay of ^{218}Po. The two-filter method is known to have a strong humidity dependence at low relative humidities, but in the range 20-90% the dependence is less critical (ThL 70). Before the two-filter tube is deployed for *in situ* measurements, it should be evaluated under different conditions of flow rate, collection time, and radon concentration to test the linearity of the system. Calibrations should also be conducted under these same conditions.

The sensitivity of the two-filter tube method depends primarily on the tube volume, sampling flow rate, and sampling and counting times. With a 1-liter tube and typical flow rates (0.1-0.2 $\text{L}\cdot\text{s}^{-1}$), sampling times (5-10 minutes) and counting times (1-6 minutes), radon concentrations on the order of 5 to 10 $\text{pCi}\cdot\text{L}^{-1}$ can be measured. With larger tubes, larger flow rates, and longer times, sensitivities as low as 0.1 to 0.01 $\text{pCi}\cdot\text{L}^{-1}$ have been reported [Bre 80; Geo 80].

IV.D.2. Time-Integral Methods

Time-integrated radon concentration measurements can be performed by large-volume sample collection or preconcentration techniques, followed by laboratory radon assays (see Section IV.A). One of the simplest large-volume sample collection methods was developed by Sill [Sil 69]. In this method, the air sample is collected over two days by pumping air into a Mylar bag at a very slow rate (typically 10 mL per minute). The field equipment consists of only a 40-L Mylar bag and a storage battery powered aquarium aerator pump with a flow regulator. The radon concentration is assayed in the laboratory by pumping the sample from the bag through a cold charcoal trap, and then transferring the radon from the charcoal to a suitable scintillation or internal gas counting device. The method is reported to be very sensitive, capable of measuring radon concentrations in the range of 0.01-0.1 $pCi \cdot L^{-1}$ [Bre 80; Geo 80]. Alternative large-volume preconcentration sampling methods include collecting the air sample by pumping the air through cooled charcoal traps [Bud 74; ISC 72], and other cryogenic techniques [HaP 80; PDH 80]. In these cases, the preconcentrated air samples are also returned to the laboratory for radon assay. The accuracy of these methods is primarily dependent on the accuracy with which the flow rate or sample volume can be measured, and the losses of radon during collection and transfer. George [Geo 80] recently summarized some of the possible sources of inaccuracy and calibration requirements for the measurement of sample volumes and flow rates.

The completely passive track-etch film techniques, which have been used for many years in uranium exploration [IAEA 76], can also be used to determine time-integrated radon concentration [Cou 76b; FAF 72; FrB 77]. Although they have been used for this latter application, the method is probably more suitable for estimating working levels and total alpha-particle incidence [Lov 69; RLN 69; Cou 76b; FrB 77]. For this method, a small piece (nominally 1 cm^2) of a suitable plastic film such as cellulose nitrate, is exposed for a long period of time in the air to be monitored. Alpha particles, incident on the film, damage the surface. By etching the film in suitable alkaline solutions (e.g., sodium hydroxide), each point of impact can be enlarged. The resultant tracks and pits are then counted with a microscope or by other electro-optical means [FrB 77] to provide a measure of the total number of alpha particles incident on the film during the exposure period. Various types of membranes and filters have been used with the films to achieve different hold-up times that can be used to exclude thoron, and to filter the particulate radon progeny radionuclides [Alt 80]. Specific track-etch methodologies for radon dosimetry have recently been reviewed by Frank and Benton [FrB 77] and will not be detailed here. In general, direct calibrations to relate track density to the quantity of interest (radon concentration, working level, etc.) must be performed for each specific application [Cou 76b]. Because of its simplicity and low cost, track-etch methods offer great potential for wide-scale deployment. Breslin [Bre 80] noted, however, that it has not been widely used because of its inaccuracy and poor sensitivity. At typical indoor radon concentrations, year-long exposure periods are needed to obtain sufficient track density; and at that concentration, the precision is poor (about ±50%). Other than unpublished claims by a commercial vendor, very few efforts [Cou 76b; FrB 77] have been made to evaluate the method for typical indoor radon concentrations.

Another passive method to measure time-integrated radon concentrations uses an instrument commonly known as a "passive environmental radon monitor" (PERM) which was developed at the DOE Environmental Measurements Laboratory [Geo 77; GeB 77; Bre 80; Geo 80]. The instrument operates on an electrostatic-collection principle which was used in previous monitors [CTD 69; WSC 75]. These earlier versions have since evolved into continuous monitors (see Section IV.D.3) and are commonly known as "Wrenn meters". The PERMS, however, are completely passive time-integrating devices which rely on a thermoluminescent detector (TLD).

For comparison, the operation of a PERM will be described in relation to the "Wrenn meter" (see Section IV.D.3). The PERM developed by the Environmental Measurements Laboratory consists of a large 1.5-L metal funnel with a paper filter sealed to the large opening (20 cm diam. at the base) and a stopper (containing a hollow electrode) inserted in the narrow opening. The "Wrenn meter" uses a more efficient hemispherical design. The PERM samples the ambient air by molecular diffusion, although some versions utilize an electric fan to draw the air in. Air enters the device and diffuses through a bed of desiccant (to eliminate a serious humidity dependence in the instrument's performance [GeB77]), and enters the sensitive volume by diffusing through a porous barrier (used to exclude the particulate radon progeny radionuclides). The PERM uses a large filter paper (Whatman #41); the "Wrenn meter" uses a polyurethane foam medium. The radon concentration in the sensitive volume gradually equilibrates with the ambient radon concentration in the surrounding atmosphere. An electrostatic field created by application of a negative potential on the hollow electrode is used to attract and collect the positively-charged ^{218}Po ions that form within the sensitive volume by decay of the radon. The hollow electrode also serves as an exit for the air in the sensitive volume. In a PERM, an alpha-particle sensitive lithium fluoride TLD chip is located at the negative electrode to detect the α-particle emitting ^{218}Po radon daughters that accumulate on the electrode. The "Wrenn meter" uses a ZnS(Ag) scintillator to detect the daughters. Early models used photographic film (read by conventional densitometry) to obtain similar time-integrated radon concentrations [CTD 69]. More recent versions of the "Wrenn meter" operate in a continuous manner by using a photomultiplier tube coupled to the scintillator, and pulse counting [WSC 75]. For the PERM, the TLD chip is removed after a suitable period of exposure (usually one or two weeks), and read on a thermoluminescent detector. The thermoluminescence of the chip (which must be corrected for gamma-radiation background with a control chip located outside the PERM) is directly proportional to, and calibrated in terms of, a time-integrated radon concentration. The PERM has a response lag of about 5 hours [GeB 77] due to the length of time needed for air to diffuse through the desiccant, but this is short in comparison to the usual length of the monitoring period.

The sensitivity of the PERM is reported to be 0.03 $pCi \cdot L^{-1}$ in a one-week exposure period [GeB 77] which is considerably less than that obtained with a continuous reading "Wrenn meter". The difference may be due to the greater detection efficiency of the ZnS(Ag) scintillator and the more efficient hemispherical shape of the "Wrenn meter" sensitive volume. .

The PERM must be directly calibrated in atmospheres having known radon concentrations [GeB 77; Geo 77], and under conditions simulating field atmospheres. This can be accomplished by placing the device in a chamber containing radon under controlled conditions. The chamber atmospheres can then be sampled, and the radon concentration measured by methods of known accuracy (e.g., with scintillation flasks or pulse ionization chambers). Alternatively, the chamber could be filled with a known amount of radon which is obtained from an equilibrated ^{226}Ra standard. Standard procedures for the performance, use, and calibration of TLD systems when used to measure environmental gamma-radiation are available [ANSI 75; PBG 76; GJP 80], but George has noted that no recommended procedures for the use of TLDs for radon monitoring are available [Geo 80]. He has also suggested, however, that the same performance specifications and testing procedures that are used for gamma-radiation applications could equally well be applied for measurements of alpha particles. In this same paper, George [Geo 80] also addressed some of the sources of inaccuracy and practical problems encountered in the use of TLDs for radon. Measurement precision for the PERM developed by the Environmental Measurements Laboratory was found during calibration experiments to be about 5% to 10% (coefficient of variation). At typical ambient levels (say 1 $pCi \cdot L^{-1}$), one can probably expect the monitor to be less precise. Considering the large correction for the gamma-radiation background and the uncertainty in presently available thermoluminescent dosimetry (TLD) techniques, the accuracy, even with a good calibration, is probably no better than 20% to 30%.

A commercial instrument similar to the PERM is available, but is reported to be much less sensitive than the original Environmental Measurement Laboratory design and is humidity dependent [Bre 80].

IV.D.3. Continuous Methods

Radon concentrations in indoor air continuously change, and, as a result, measurements based on a single grab sample are rarely sufficient [Bre 80]. For this reason, continuous methods are probably more useful. Obviously, any of the "instantaneous" methods described in Section IV.D.1. could be performed repeatedly to obtain a continuous record, but methods designed specifically for continuous monitoring are considerably more convenient. Many of them are adaptations of previously described methods that were modified for use as continuously-reading monitors. The three most common are: continuously-reading flow-through devices using either an ionization chamber [WaH 69] or a scintillation flask [HLW 57] (see Sections IV.A. and IV.D.1.); an automated two-filter tube method [Tho 77a] (see Section IV.D.1.); and the diffusion-electrostatic "Wrenn meter" [WSC 75] (see Section IV.D.2.).

Flow-through ionization chambers have been used for many years to measure radon concentrations in uranium-mine atmospheres, but their use indoors has been very limited. A typical detector consists of a gamma-radiation compensated, flow-through ionization chamber, of approximately 1-L volume, preceded by a filter and electrostatic precipitator. All attached and unattached radon progeny radionuclides are removed from the air stream entering the chamber so that the ionization current is due only to radon decay. The units are also equipped with a pump to draw air through the filter, precipitator and chamber; an electrometer to measure the ionization current; and some suitable recording device. A calibration procedure for this type of instrument (using an NBS ^{226}Ra solution standard as a source of radon) was described by Waters and Howard [WaH 69]. They reported a full-scale sensitivity of 66 $pCi \cdot L^{-1}$ on the most sensitive range of the electrometer, and an ability to measure radon concentrations in the 5-10 $pCi \cdot L^{-1}$ range. The estimated precision of the measurements at full-scale (i.e., 66 $pCi \cdot L^{-1}$) was approximately 10% [WaH 69].

Flow-through scintillation flasks were first developed in the mid-1950s by the former Health and Safety Laboratory (now Environmental Measurements Laboratory) [HLW 57], and have become much more popular than ionization chambers for measuring indoor radon concentrations. Other groups have since reported the development of similar instruments, and a commercial version is available. Breslin [Bre 80] and George [Geo 80] reported that the approximate sensitivity for this type of instrument is typically 0.5 $pCi \cdot L^{-1}$, but is dependent on the size of the scintillation flask, the flow rate, and the counting time interval.

The instrument consists of a Lucas-type scintillation flask (see Sections IV.A.2. and IV.D.1.) with two ports that allow air to be continuously drawn through the flask. It is permanently coupled to a photomultiplier tube and pulse counter, and thus can continuously monitor changes in the radon concentration in the air drawn through the flask. As noted by Breslin [Bre 80], the fallacy in this arrangement is that even if the inlet stream is filtered, radon progeny radionuclides produced within the flask "plate out" and deposit within the detecting volume. As a result, the count rate is affected by the plated-out progeny that arise from radon sampled in the previous several hours [Tho 77a; BVP 79]. This problem persisted for many years until a procedure, first proposed by Thomas [Tho 77b], was developed by Countess and Thomas [CoT 77] to correct for the previously-deposited radon progeny. The procedure consists of an experimental determination of a "deposition constant" for the flask, and radioactive decay corrections for the previously deposited progeny radionuclides.

The continuous scintillation-flask monitor must be calibrated under the same prevailing conditions and in the same manner as it is used for field measurements [GeO 80], and must include the correction for the previously deposited radon progeny. The precision of the calibration factors for individual flasks is about ±5% [Tho 77a], presumably due to the variability in the scintillator coating on the walls. The overall precision, including "counting errors", is estimated to be about ±20% (coefficient of variation) for a radon concentration of 1 $pCi \cdot L^{-1}$ [Tho77a].

Another type of instrument for continuously measuring indoor radon concentrations is an automated two-filter tube monitor [Tho 77a]. This instrument consists of a two-filter tube (see Section IV.D.1.) with a scintillation detector positioned in front of the exit filter to continuously detect the radon progeny. Like the continuous scintillation-flask monitor, this instrument is also susceptible to interference from previously deposited radon progeny. Similar corrections for the radionuclides deposited during previous sampling intervals must be made. Another source of inaccuracy with continuous two-filter tubes arises from the collection of the longer-lived thoron progeny radionuclides on the exit filter. This has been eliminated from some instruments by passing the air through two large delay chambers (850 L each) located upstream from the two-filter tube [Tho 77a; Geo 80]. At a flow rate of 5 $L \cdot s^{-1}$, the chambers provide a delay time of about six minutes which is sufficient to allow decay of the short-lived thoron (55 s halflife) [Tho 77a]. These conditions have little effect on the radon, but remove about 98% of the thoron. Use of delay chambers is recommended whenever the thoron concentration is suspected to be approximately equal to or greater than the radon concentration [Tho 77a].

A more recent modification of the two-filter tube method uses an automatic drive that changes the exit filter every hour and transports the filter to a built-in scintillation detector [Neg 79]. This modification eliminates the accumulation of thoron progeny (thus obviating the need for delay chambers), and the need for corrections for previously-deposited progeny.

As with continuous scintillation flasks, automatic two-filter tubes must be directly calibrated in the same manner and for similar conditions used in field measurements [Tho 77a; Geo 80]. The overall precision at 1 $pCi \cdot L^{-1}$ is reported to be similar (i.e., an approximate coefficient of variation of about ±20%) [Tho 77a]. Negro [Neg 79] directly compared results from the automatic-drive two-filter tube monitor to the average of grab samples assayed with pulse ionization chambers (see Section IV.A.1.). For radon concentrations in the range 0.1-0.2 $pCi \cdot L^{-1}$, the differences between the monitor reading and grab-sample averages were less than 10%. The sensitivity of continuous two-filter tube monitors depends on the volume of the tube, and sensitivities of 0.01 $pCi \cdot L^{-1}$ have been reported [Bre 80]. Very large volume tubes are required to achieve this sensitivity, however, and sensitivities on the order of a few tenths of a picocurie per liter are more typical for portable units.

Most continuous monitors based on a diffusion and electrostatic-collection technique are adaptations of an instrument that is popularly called a "Wrenn Meter" [WSC 75; PDH 80]. As described in Section IV.D.2., the instrument is based on the diffusion of radon through a porous barrier into a hemispherical sensitive volume, and the attraction and collection of the ^{218}Po radon daughters by an electrostatic field applied within the sensitive volume (typically one liter). In the original design by Wrenn, et al. [WSC 75], the central electrode consisted of a hemispherical lucite light pipe painted with a ZnS(Ag) scintillator coating which was then covered with aluminized mylar to obtain an electrically conductive surface. Scintillations produced in the ZnS(Ag) coating are detected by a photomultiplier tube which is optically coupled to the light pipe.

As in the other continuous monitors, interferences from previously deposited radon progeny partially mask changes in the ambient radon concentration. Although corrections similar to those used for other monitors could, in principle, be applied, it does not appear that such corrections are presently being made [Bre 80; Geo 80]. Their absence seriously compromises the utility of these instruments for accurate measurements of radon concentrations. Another limiting deficiency may be the humidity dependence of these instruments. Although Wrenn, et al. [WSC 75] reported that the collection efficiency "appears to be unaffected by normal fluctuations of relative humidity", Breslin [Bre 80] and George [Geo 80] suggested that these instruments have a serious humidity dependence. Tests at the Environmental Measurements Laboratory with similar monitors showed a 6% decrease in sensitivity for a 10% increase in relative humidity [New 78]. This apparent discrepancy may be due, in part, to changes in the shape and size of the original "Wrenn meter".

As for other continuous monitors, instruments based on the electrostatic-collection technique must also be directly calibrated by exposure to known radon concentrations [WSC 75; NeW 78; Geo 80]. The sensitivity of continuous "Wrenn meters" is in the range of 0.1-0.5 $pCi \cdot L^{-1}$ [WSC 75; PDH 80].

A similar ten-liter instrument which uses a solid-state detector rather than a scintillation detector to detect the alpha particles from the radon progeny has been investigated [NeW 78]. This instrument can be used to separately resolve the ^{218}Po and ^{214}Po alpha particles. It is reported that, by detecting only the alpha particles from the shorter-lived ^{218}Po daughters, the instrument's response to changes in radon concentration is ten times faster than other instruments [NeW 78].

IV.E. Conclusions and Recommendations

As suggested throughout this chapter, there is a plethoric number and variety of measurement methodologies which have been used for both laboratory and in situ studies of radon transport and exhalation. Although some of these measurement methods, particularly the laboratory-based ones, have survived the test of time, others have never undergone a detailed assessment and evaluation. The need to evaluate the suitability of the track-etch technique for measuring indoor radon concentrations is perhaps the most representative example. Although several fragmentary studies of the adequacy of the track-etch techniques have been or are being performed, a major unequivocal assessment of the suitability and reliability of the method needs to be performed. Similarly, some of the radon flux density and continuous monitoring methods would also benefit from more broad-based, critical assessments and evaluations. As noted earlier, this need for critical assessments of measurement methodologies has also been recently identified by the Radiation Policy Council Task Force on Radon in Structures [RPC 80]. In fact, the need to give more consideration to the evaluation of methods prior to their wide-spread deployment is pleaded whenever three or more "radon experts" gather in the same room!

Equally important, is the continuing and often cited need for the development of uniform measurement procedures, the development of calibration standards and facilities, and the development of mechanisms for measurement assurance and interlaboratory comparison programs that can be related to national measurement standards. At present, the only national standards that are available for any radon-measurement-related quantity are NBS ^{226}Ra solution standard reference materials which are used for radon analyses. Although some laboratories, such as the DOE Environmental Measurements Laboratory, have over many years striven to relate all of their radon-related measurements to this standard, not all users can be expected to have either the resources or capability to accomplish this. Many users now need and will continue to need readily available measurement standards, calibration services, and measurement assurance mechanisms.

The importance of measurements to understanding radon transport and exhalation from building materials, and to future remedial corrections and controls of this source of indoor radon cannot be overstated. Yet, this importance is overshadowed by the absence of some of the most basic measurement support services. Inasmuch as NBS is the only national organization which has both the responsibility and long-term commitment to insuring the quality of measurements, it is recommended that NBS be encouraged and supported in undertaking studies to assess and evaluate the adequacy of the more important measurement methodologies, and in developing measurement standards, calibration facilities and measurement assurance mechanisms that can be disseminated to the entire radon-measurement community.

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NBS-114A (REV. 2-80) PB82 112384

U.S. DEPT. OF COMM.
BIBLIOGRAPHIC DATA SHEET *(See instructions)*

1. PUBLICATION OR REPORT NO.
NBS TN 1139

2. Performing Organ. Report No.

3. Publication Date
September 1981

4. TITLE AND SUBTITLE

Radon Transport Through and Exhalation From Building Materials: A Review and Assessment

5. AUTHOR(S)

R. Collé, R.J. Rubin, L.I. Knab, and J.M.R. Hutchinson

6. PERFORMING ORGANIZATION *(If joint or other than NBS, see instructions)*

NATIONAL BUREAU OF STANDARDS
DEPARTMENT OF COMMERCE
WASHINGTON, D.C. 20234

7. Contract/Grant No.
AD-13-F-0-133-0

8. Type of Report & Period Covered
Interim

9. SPONSORING ORGANIZATION NAME AND COMPLETE ADDRESS *(Street, City, State, ZIP)*

Office of Radiation Programs
U.S. Environmental Protection Agency
Washington, DC 20460

10. SUPPLEMENTARY NOTES

☐ Document describes a computer program; SF-185, FIPS Software Summary, is attached.

11. ABSTRACT *(A 200-word or less factual summary of most significant information. If document includes a significant bibliography or literature survey, mention it here)*

This report was prepared, at the request of the U.S. Environmental Protection Agency, for the purpose of reviewing, assessing, and summarizing what is currently known about radon transport through and exhalation from building materials. In four chapters, the report 1) considers the routes of entry of radon into buildings, describes the basic models for radon transport through building materials, critically reviews the small number of existing values for the necessary transport coefficients, and summarizes the solutions of both steady-state and time-dependent transport cases; 2) reviews and considers how the microstructural properties and internal characteristics of building materials may affect the transport and exhalation of radon; 3) considers the exhalation process from a more macroscopic, phenomenological viewpoint, and summarizes selected experimental data on radium concentrations in building materials, radon flux and exhalation from soils and building materials, and the effects of meteorological variables on radon exhalation; and 4) reviews and assesses various measurement methodologies that are used for laboratory and in situ studies of radon transport and exhalation. Needs for further research in each area are also recommended.

12. KEY WORDS *(Six to twelve entries; alphabetical order; capitalize only proper names; and separate key words by semicolons)*

buildings; concrete; diffusion; exhalation; materials; measurement; permeability; radon; transport

13. AVAILABILITY

☒ Unlimited
☐ For Official Distribution. Do Not Release to NTIS
☒ Order From Superintendent of Documents, U.S. Government Printing Office, Washington, D.C. 20402.
☐ Order From National Technical Information Service (NTIS), Springfield, VA. 22161

14. NO. OF PRINTED PAGES
101

15. Price
$4.50

USCOMM-DC 6043-P80

U.S. GOVERNMENT PRINTING OFFICE: 1981-340-997/1799

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